

Chromium Contaminated Soil-Groundwater Systems at COPR Site in Rania and Khan Chandpur Villages, Kanpur Dehat, India

1. INTRODUCTION

Observing the increasing concern over groundwater contamination and deteriorating soil-water quality, we moved to Rania and Khan Chandpur villages in Kanpur Dehat district in Uttar Pradesh. Rania-Khan Chandpur site is one of the highly contaminated sites in India (Prakash et al. 2011; CPCB, 2023) due to Chromite Ore Processing Residue (COPR), which is a hazardous (waste) remnant of the chromate (CrO_4^{2-}) extraction roasting process (Földi et al. 2013; Matern et al. 2017; 2020). As per report of the oversight committee, Hon'ble NGT, U.P. Lucknow, there is an estimated volume of 1,22,800 m³ COPR at Rania site (which has recently relocated to the treatment, storage, and disposal facility (TSDF) located 25Km from this site near Akbarpur, Kanpur Dehat). Both the villages are settled in the vicinity of the leather processing clusters and COPR waste from these tanneries and related industries has severely contaminated soils, the surface-water, and the groundwater. The residents are drinking Cr-containing groundwater and are continuously at risk through the poisonous intake either through groundwater or locally grown vegetables or other sources (Gupta et al. In Preparation). Regardless of this situation, the remediation practices of Cr-contaminated soil-water systems remain unattempted at the Rania-Khan Chandpur, this is because the site-scale Cr fate and mobility in the subsurface is still less investigated.

Cr leachate from the COPR can infiltrate in the vadose zone and ultimately reach the (shallow-) groundwater which can lead to huge Cr contamination in the subsurface. Our initial (Gupta et al. 2023a; Deoli et al. 2023) and other's works (Singh et. al., 2009; Matern et. al., 2017) have found that groundwater, surface-water, and soil samples from **the past** have shown signs of a Cr emanating from the COPR dump site in and around Rania-Khan Chandpur villages. In the subsurface, Cr can be in the form of Cr (III) or Cr (VI), the mechanisms and kinetics of Cr release are obscure (Bhattacharya et. al., 2019). The release of Cr from the COPR dump site can be influenced by factors such as the pH of the water/soil, the presence of other chemicals or ions, the hydrogeology and hydrology of the site, the above and below-ground ecology and microbial community dynamics, and so on. Further, climate change may influence the fate and mobility of the Cr by altering the hydro-metrolological and subsurface conditions. A better understanding is needed to improve remediation practices including evaluation of potential Cr transport pathways and rates, and more efficient clean-up strategies. However, there remains a dearth of knowledge on Cr fate, transport, and remediation in the soil-water system.

Further, vegetation and microbial communities were identified that can thrive in Cr stress conditions. Plant-bacterial relationships can adsorb, decrease, and precipitate Cr, especially when

37 it comes to Cr (III) and Cr (VI) forms (Thatoi et al. 2014). These plants exhibit strong potential for
38 stabilizing and detoxifying contaminated soils because of their inherent resistance mechanisms
39 and high metal-uptake capacities (Antoniadis et al. 2017). In addition to these plants, several
40 microbes exhibit remarkable abilities to reduce Cr concentrations via the sulphate transport
41 pathway, passive transport, and chromium accumulation pathways (Wang et al. 2015). Among the
42 widely used techniques is the sulphate transport system, which is found in many bacteria, including
43 *Salmonella typhimurium*, *Bacillus subtilis*, *Escherichia coli*, *Pseudomonas fluorescens*, and
44 *Alcaligenes eutrophus* (Su et al. 2023). The function of sulphate transporters is essential for Cr
45 (VI) tolerance because they control both sulphur availability and chromium uptake. This is because
46 CrO_4^{2-} and sulphate (SO_4^{2-}) have similar chemical makeups and can compete for the same
47 transporters (Takahashi et al. 2012). Hence, the most effective method for removing Cr (VI) from
48 the hydrogeological matrix is the reduction of Cr (VI) to Cr (III) followed by its adsorption (Fan
49 et al. 2019).

50 Critical levels of Cr contamination have been found in the groundwater of Rania-Khan
51 Chandpur villages. This contamination primarily results from the improper disposal of tannery
52 waste, including Basic Chrome Sulphate (BCS) sludge. Groundwater has been reported to contain
53 Cr(VI) concentrations as high as 34.8 mg L^{-1} (Matern et al. 2017), and total chromium levels as
54 high as 40 mg L^{-1} (Gupta et al. 2024), far surpassing the 0.05 mg L^{-1} Bureau of Indian Standards
55 (BIS) permitted limit for drinking water. Significant groundwater pollution is additionally
56 indicated by the high levels of electrical conductivity ($2243 \mu\text{S cm}^{-1}$), sulphate (385 mg L^{-1}), and
57 fluoride up to 4.02 mg L^{-1} (Srivastava et al. 2013). According to health risk assessments, children's
58 non-carcinogenic Hazard Index (HI) can be as high as 5.214, which is far higher than the safe
59 threshold ($\text{HI} > 1$) and indicates negative health impacts. Furthermore, all receptors have a
60 carcinogenic risk for Cr (VI) greater than 1×10^{-6} , and children are at risk for cancer almost 8.6
61 times higher than adults (Guleria et al. 2022).

62 A comprehensive analysis of soil samples from contaminated areas can reveal a diverse
63 array of bacterial communities in both the soil and COPR samples. These findings can offer
64 valuable insights into the microbial dynamics of subsurfaces affected by Cr contamination and
65 underscore the significant Cr retention potential of bacteria (Tripathi et al. 2022). A better
66 understanding of native plants and microbes will help to design bioremediation of such a highly
67 contaminated site. This integrated approach of using both native plants and microbes holds
68 promise for sustainable and effective bioremediation strategies. But to provide optimal
69 hydrogeological conditions for remediation is equally important.

70 Cr contamination, particularly from COPR, has caused significant environmental damage,
71 especially in places like the Rania-Khan Chandpur. The vadose zone and groundwater have been
72 exposed to chromium, mostly in Cr (VI), which has leached from COPR disposal sites and resulted
73 in higher than usual amounts of this hazardous metal in water sources. The region's humid
74 subtropical climate enhances the mobility of Cr(VI), thereby increasing contamination levels and

75 posing serious risks to human health, plant life, and soil microbial communities. A comprehensive
76 analysis of soil samples from contaminated areas can reveal a diverse array of bacterial
77 communities in both the soil and COPR samples. These findings can offer valuable insights into
78 the microbial dynamics of the subsurface affected by Cr contamination and underscore bacteria's
79 significant Cr retention potential. A better understanding of native plants and microbes will help
80 to design Cr-bioremediation for such a highly contaminated site. This integrated approach of using
81 native plants and microbes holds promise for sustainable and effective bioremediation strategies.
82 However, to provide optimal hydrogeological conditions for remediation is equally important.
83 Previous studies have often lacked the use of UAV surveys to accurately map contaminated zones
84 and assess the land-use changes caused by Cr contamination. This paper integrates UAV surveys
85 with groundwater and soil data for a more comprehensive understanding of contamination spread
86 and provides insights into the spatial extent of chromium distribution.

87 Previous studies have found that the use of UAV surveys to accurately map contaminated
88 areas and assess changes in land use caused by chromium pollution has often been overlooked
89 (Khosravi et al. 2021; Capolupo et al. 2015). This research has provided valuable information
90 about the extent of Cr distribution in space and has integrated UAV surveys with groundwater and
91 soil data to gain a more comprehensive understanding of the contaminated areas. To gain a more
92 comprehensive understanding of contamination spread, this work will integrate UAV surveys with
93 groundwater and soil data. Furthermore, it will provide insight into the spatial extent of Cr
94 distribution.

95 Considering the research gaps prior to the implementation of this research work, a series
96 of investigations has been initiated to generate baseline data of soil-water quality at the Rania-
97 Khan Chandpur site. This has resulted in basic knowledge of the initial site conditions and
98 profusion of excellent baseline data facilitating a better understanding of the soil-water system at
99 this site. In this research work, the fate and mobility of Cr in the subsurface under varying
100 hydrogeological conditions will be investigated first. Thereafter, the performance of the
101 remediation techniques will be evaluated for restoration of Cr-contaminated soil-water systems at
102 a demonstrative site in Rania-Khan Chandpur villages. This project will provide very accurate,
103 multimethod techniques for the forecasting of the Cr mobility in the subsurface, which can assist
104 the site managers/stakeholders for the future site scale planning. Further, this project will not only
105 improve our understanding to manage a highly contaminated site in India, but also help in the
106 management of such contaminated sites in other parts of the globe.

107 Further, the presence of naturally resistant microbial communities makes it difficult to
108 develop effective remediation strategies for Cr contamination. Although some data exists, there is
109 inadequate documentation of the distribution and concentration of both Cr (III) and Cr (VI) in
110 various environmental media such as soil and groundwater. Despite awareness of the
111 contamination, there has been limited initiation and study of active remediation in soil-water
112 systems. In the present study our objectives are: (1) To measure Cr contamination in soil and

113 groundwater, using UAV surveys to map and prioritize remediation areas., (2) To identify native
114 plants and microbes for bioremediation, and implement a pilot project to test their effectiveness.
115 (3) To integrate hydrogeological and microbial data into a framework for long term Cr
116 management.

117 **2. Cr Global and National Distribution**

118 **2.1 Global Sites**

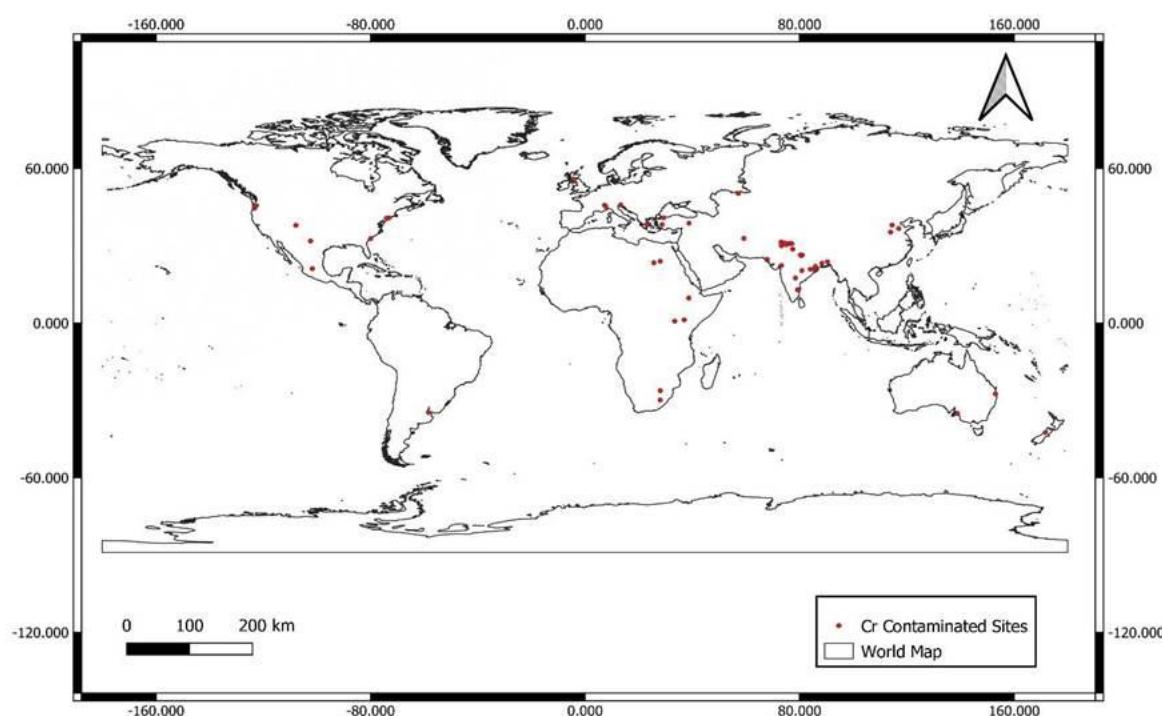
119 About ten million tons of chromium are produced globally each year. South Africa, Turkey,
120 China, India, and Kazakhstan are the top five countries for chromium production in the world from
121 which South Africa produces almost two-fifths of the chromite ores and concentrates. (Mohanty
122 et al., 2011; Mishra and Bharagave 2016). Oruko et al. (20121) investigated two industrial sites
123 Dongo Bonde Ltd. in Kenya and Bath Ore in South Africa. They found the mean Cr (VI)
124 concentration in treated effluent were 1.82 ppm and 7.9 ppm respectively. In California, the Cr
125 concentration levels in soil samples have varied between 140 to 2220 mg/kg soil (Mills et al.,
126 2011) and in groundwater samples the maximum Cr concentration level has been reported 50 µg/l
127 (Izbicki et al., 2018). Approximately 40% of water resources are polluted with Cr (VI) in California
128 (Sedman et al., 2006). In the Sarigkiol Basin of Greece, up to 120 µg/l of Cr (VI) has been observed
129 in groundwater and the Cr (VI) level varied from 20 – 55 µg/l in spring water (Kazakis et al.,
130 2017). In Sohag Governorate, Egypt, the total Cr concentration level has been reported to be up to
131 260.6 µg/l which is very higher than the preferred limit of 50 µg/l, set by WHO 2011. The Cr (VI)
132 variation in that area varies between 12 to 58 µg/l (Salman et al., 2020). In Aosta Italy, the Cr (VI)
133 in groundwater varies maximum up to 165 µg/l from minimum value of 0.09 µg/l.

134 Countries like USA, Argentina, Pakistan, Bangladesh, Germany, Indonesia have suffered
135 from Cr concentration in groundwater (Table 1; Figure 1) (Coyote et al., 2020; Ali et al., 2022;
136 Khan et al., 2022; Vainshtein et al., 2003; Astuti et al., 2023). As per the study conducted in Henan
137 Province (Chen et al., 2021), the concentration of Cr (VI) was maximum up to 299.99 mg/l in
138 shallow groundwater samples and in deep groundwater samples it varies up to 56.30 mg/l. High
139 concentrations of Cr and Cr (VI) have also been reported in some belts of Turkey, Brazil, Morocco
140 (Tokatli, 2021; Hirata et al., 2007; Lotfi et al., 2020). Due to Cr concentration in soil-water system,
141 stomach and lung cancer have been reported in Jin Zhou, China (Zhang and Li, 1987).

142 **2.2 Indian Sites**

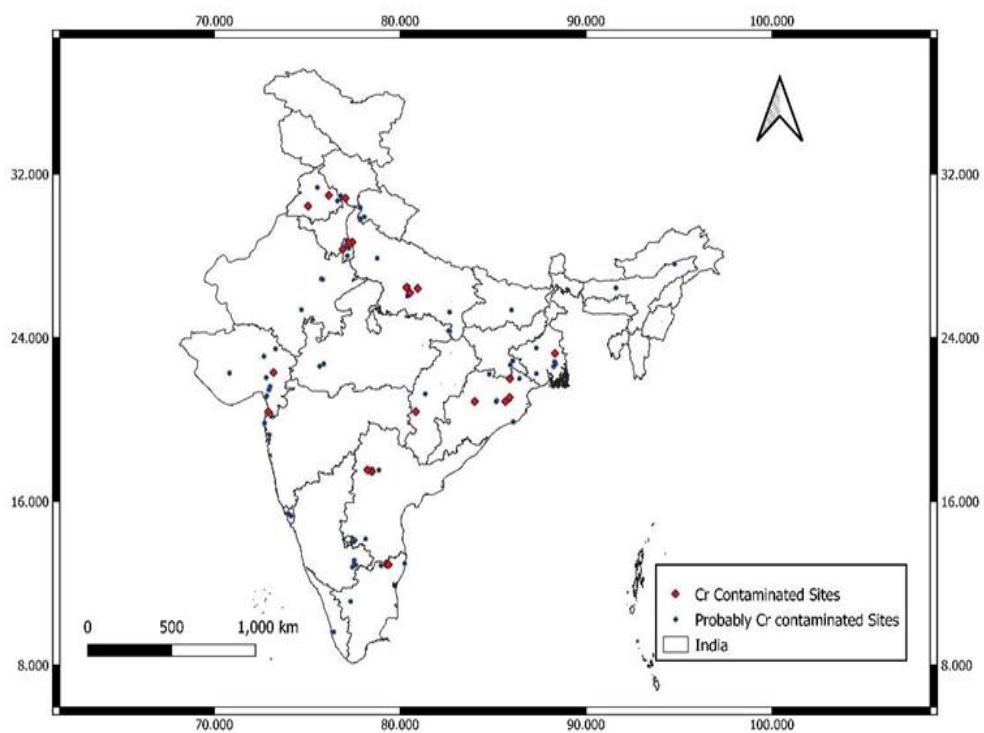
143 In India, there are more than 18 chromium contaminated sites and total of 111 probably
144 Cr contaminated sites including Rania-Khan Chandpur (Kanpur, Uttar Pradesh), Sukinda Valley
145 (Odisha) and Ranipat (Tamil Nadu) are the worst-hit states with a high abundance of Cr in soil-
146 water system (CPCB, 2023) (Table 2; Figure 2). In many other states, the levels of Cr have
147 exceeded the permissible limit which creates the consequences. The highest concentration of Cr

148 and Cr (VI) have been reported in Sukinda Valley with values of 48.7–250.2 and 21.4–115.2 $\mu\text{g/l}$
149 respectively (Naz et al., 2016). In Rania-Khan Chandpur Villages of Kanpur Dehat, Uttar Pradesh
150 The maximum value of Cr in groundwater reported as 33.88 mg/l (Singh et al., 2012). Matern et.
151 al. (2017) found the maximum Cr concentration in Rania and Chhiwali (Kanpur Dehat Uttar
152 Pradesh) maximum up to 34.8 mg/l and 115 mg/l respectively. In Bangalore India, the Cr (III)
153 concentration in surface soils very from 14.9 mg/kg to 718.1 mg/kg whereas in the deep soil 15.1
154 to 116.1 mg/kg which is showing the surface Cr pollution load is more due to the COPR dumping
155 (Ratnalu ei al., 2021). Previously, similar study has been carried out by Paul et al. (2014) for
156 Jajmau India for agriculture soil, which contains 40,500 mg/kg Cr and 1,400 mg/kg Cr (VI). The
157 groundwater table of Punjab, Rajasthan, New Delhi have also been reported to Cr contaminated.



158
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Figure 1: Cr contaminated sites of World



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161

Figure 2: Cr contaminated and probably Cr contaminated sites of India

Table 1: Global distribution of Cr in different subsurface soil conditions, Vadose Zone and in groundwater systems

Citation	Lat/long	Scale of Study	Samples/Environmental Conditions	Methods	Key Observations
Szecsody et al. (2019)	Hanford (36.327728N, 119.645912W)	Lab Scale	Cr contaminated sediment collected from vadose zone	<ul style="list-style-type: none"> • 1D leaching experiment in 2.35 cm diameter and 15-20 cm long column. • Cr (VI) was measured via a spectrophotometer at 540 nm. • EDS was analyzed for Cr in sediment. • Batch Experiment 	<ul style="list-style-type: none"> • Columns with high concentrations of Cr (VI) (up to 187 mg/l Cr (VI)) rapidly leached more than 90% of the Cr (VI) in the vadose zone. • In natural groundwater Barium was not present above the detection limit for the studied area. • A significant amount of Cr was found in the surface phase identification of vadose zone sediments
Ballesteros et al. (2017)	Ltitlan (21.127800 N, 98.650787W)	Lab Scale	Soil, Slag and Demolition residues chromates samples were collected from the dumping site.	<ul style="list-style-type: none"> • XRF, atomic absorption ICP and C and S combustion were used for chemical characterization. • TCLP was used for the lixiviation of the toxic substance. 	<ul style="list-style-type: none"> • All Cr (VI) were reduced to Cr (III) in the residual glass network. • The Cr (VI) in the glass ceramics was low (<0.22 mg/kg). • The final ceramics showed adequate mechanical properties.
Broadway et al. (2010)	Glassgow (55.8617° N, 4.2583° W)	Lab Scale	<ul style="list-style-type: none"> • 27 soil samples collected. 	<ul style="list-style-type: none"> • Chromium was determined by ICP-OES. 	<ul style="list-style-type: none"> • Total Cr and Cr (VI) in the soil were 3680 mg/kg and 1485 mg/kg respectively. • In a lung inhalation simulation, it was observed that the fine

				<ul style="list-style-type: none"> • UBM and Gamble's solution used for speciation of Cr. 	<p>fraction of soil (with particles smaller than 10 µm) from specific Cr (VI)-contaminated sites contained bioaccessible forms of chromium.</p>
Ceballos et al. (2023)	Glasgow (55.8617° N, 4.2583° W)	Lab Scale	16 soil samples were collected	<ul style="list-style-type: none"> • Sequential extraction process (SEP) used for characterizing the total Cr. • Flow through experiment and 1 column experiment were performed. The diameter and length of the column were 2 cm and 3 cm respectively. • (1D) reactive transport modelling using the CrunchFlow code. 	<p>Cr (VI) was released from dissolving Cr (VI)-rich phases of the solid waste layer.</p> <ul style="list-style-type: none"> • The soil profile, Cr predominantly exists in association with S, Fe, K, and Al, forming components such as Cr (VI)-rich jarosite, Cr (VI)-rich ettringite, and Cr (VI)-bearing hydrocalumite.
Panagiota kis et al. (2015)	Thiva Greece (38.3226° N, 23.3204° E)	Lab scale	61 water samples were collected from wells. 14 samples were collected.	<ul style="list-style-type: none"> • Water quality tests have been done by standard method. • XRD has been used for mineralogical analysis. • Two samples were analyzed for XANES. 	<p>Cr is present in topsoil of both areas due to the ultramafic geologic background of the entire region.</p> <ul style="list-style-type: none"> • The origin of Cr (VI) detected in groundwater of the wider area of Thiva is primarily of geogenic origin. • The Cr VI in tested two groundwater samples were Cr

					(VI) groundwater plumes of 160 µ g/L and 75 µ g/L.
Yan et al. (2022)	Shaanxi (34.2649° N, 108.9542° E)	Lab Scale	A total of 75 groundwater samples collected. 265 sediment samples collected.	<ul style="list-style-type: none"> Standard water quality methods have been used for chemical analysis of water. ICP-AES has been utilized for major cations and ICP-MS was used for trace elements. XRF was used for solid samples analyzed. 	<ul style="list-style-type: none"> Deep aquifer groundwater had a relatively high median Cr concentration of 142 µg/L, whereas shallow groundwater exhibited a lower median Cr concentration of 33.8 µg/L. In deep aquifers had anomalously high contents of total Cr (average 115 mg/kg). Relatively high pH and ionic strength mobilized the adsorbed Cr (VI) into groundwater. Manganese oxides were the main oxidant for Cr (III) oxidation into Cr (VI) in the aquifers.
Ceballos and Bea (2018)	Aguel (13.3319° N, 39.5139° E)	Field and Lab Scale	<ul style="list-style-type: none"> 7 Groundwater samples have been collected. Average rainfall recorded was 1008 mm/year. Average annual recharge is 133 mm/year. 	<ul style="list-style-type: none"> Standard water quality methods have been used for chemical analysis of water. Reactive transport modelling by the MIN3P UQ analysis 	<ul style="list-style-type: none"> The concentrations of Cr (VI) in both aquifers are influenced by their hydraulic connectivity. The Cr concentration varies between 0.001 to 5.63. A surface-based chromium (Cr) source term is the most realistic scenario to account for observed Cr concentrations.

				<ul style="list-style-type: none"> • Cr (VI) concentrations in the PUAQ are sensitive to organic matter contents.
Homa et al. (2016)	Ethiopia Tannery Share Company (8°27.154' N and 39+03.894' E	Lab Scale	<ul style="list-style-type: none"> • Water samples, Vegetable (cabbage, green pepper, tomatoes, and onions) samples and soil samples. • Average annual rainfall of 800 mm, and minimum and maximum temperature of 17.5°C and 26°C 	<ul style="list-style-type: none"> • Cr determination has been done using AAS. • One way ANOVA has been used for statiscal analysis. • The total chromium concentrations vary, ranging from 10.85 ± 0.885 mg/L to 39.696 ± 0.326 mg/L in water, 16.225 ± 0.12 mg/Kg to 1581.667 ± 0.122 mg/Kg in agricultural soil, and 1.0758 ± 0.05348 mg/Kg to 11.75 ± 0.206 mg/Kg in vegetable samples. The highest levels of chromium (VI) found from the speciation study were 2.23 ± 0.032 mg/Kg and 0.322 ± 0.07mg/L in soil and water. • The highest concentration of Cr (VI) was detected in onion root (0.048 ± 0.065 mg/Kg) among vegetables.

164 **Table 2: Cr distribution across different Indian states.**

S. No.	State	Place	Type of Samples	Min Value (mg/l)	Max value (mg/l)	References
1	Uttar Pradesh	Rania-Khan Chandpur	Groundwater	4.65	33.88	Singh et al. (2012)
2	Tamil Nadu	Ranipat	Borewells, open wells and handpumps	0.01	40.52	Vijayakumar et al. (2022)
3	Tamil Nadu	Vellore	Groundwater	0	0.0286	Venkatesan et al. (2020)
4	Tamil Nadu	Vellore	Vegetation	0.02	0.70	Venkatesan et al. (2020)
5	Tamil Nadu	Chromepet, Chennai	Groundwater	0.004	0.99	Brindha and Elango (2012)
6	Arunachal Pradesh	Papum Pare	Tube wells	0.14	0.18	Goswami et al. (2023)
7	Orrisa	Sukinda	Mine water, groundwater, and surface water	0.04	5.2	Equeenuddin et al. (2020)
8	Punjab	Sutlej River sub-basin	Groundwater and surface water	0	85.2	Krishan et al. (2021)
9	Rajasthan	Bhiwadi	Groundwater	0.001	0.021	Rajput et al. (2020)
10	Assam	Dhemaji	tubewells and ringwells	0	0.31	Buragohain et al. (2009)

11	Tamil Nadu	Kancheepuram	Open wells and Bore wells	0.25	0.9	Balakrishnan et al. (2008)
12	Chhattisgarh	Nadini-Khundini village of the Durg district	Groundwater	0	22	Banchhor et al., (2020)
13	Punjab	Malwa	Groundwater	0.001	3.6	Sharma and Dutta (2017)
14	Chhattisgarh	Bhilai-Durg	Bore wells	0	12.3	Banchhor et al., (2020)
15	Uttar Pradesh	Jhansi	Groundwater	0.042	0.061	Verma et al. (2016)
16	Uttar Pradesh	Jajmu	Soil	3.4 mg/kg	40529	Paul et al. (2015)
17	Karnataka	Peenya industrial area, Bengaluru	Soil	14.9	718.1	Ratnalu et al. (2021)
18	Orissa	Jajpur	Tube well and surface water	0	2.48	Naz et al. (2016)
19	Himachal Pradesh	Solan	Hand Pumps and Biwadi	0.61	1.07	Singh and Sharma (2020)
20	Gujrat	Surat	Soil samples	100.4	305.2	Krishna and Govil (2004)
21	Telangana	Nirmal Province	Soil	55.9	135.8	Adimalla et al. (2021)
22	Uttar Pradesh	Ghaziabad	Soil	356	2300	Chabukdhara, and Nema (2013)
23	West Bengal	Ranaghat– Fulia	Sediment soil	1.5	451	Sanyal et al. (2015)

24	Assam	Guwahati	Soil	60.1	730.7	Mahanta and Bhattacharyya (2011)
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166 **3. Hydrogeological Perspectives of Chromium Contamination**

167 **3.1 Adsorption**

168 The adsorption of a solute onto any aquifer material results in a reduction of concentration
 169 in the aqueous phase and also a retardation of the velocity of contaminant movement. Adsorption
 170 is a phenomenon where the accumulation and concentration of molecules at the interface of a
 171 sorbent material (Toth 2002). During adsorption, molecules move from the bulk phase and adhere
 172 to the porous surface in a semi-liquid state, and this process is primarily driven by the
 173 concentration-to-solubility ratio of the compound. The adsorption behaviour of Cr, including its
 174 prevalent oxidation states, Cr (III) and Cr (VI), is influenced by soil-groundwater parameters, such
 175 as pH, texture, temperature, moisture content, organic matter content, mineralogy, and cation
 176 exchange capacity (Suzuki, 1990).

177 Adsorption kinetics, isotherms, and thermodynamics are three fundamental components of
 178 adsorption experiments, important for understanding the removal of Cr from soil-groundwater
 179 systems (Table 3). The kinetic study of the adsorption process focuses on elucidating the
 180 adsorption rate of Cr (VI) by the adsorbent, including the study of different mechanisms and factors
 181 governing the rate at which chromium ions adhere to the solid phase. In the adsorption of Cr, an
 182 initial fast adsorption rate is observed, but it gradually decreases over time until it achieves
 183 equilibrium.

184 To understand and analyse the adsorption process of Cr (VI), the adsorption kinetic models,
 185 Boyd external diffusion, pseudo-first order (PFO), Weber and Morris internal diffusion, pseudo–
 186 second order (PSO), second-order rate equation and Elovich kinetic models have been widely used
 187 (Chien et al., 1980; Gupta and Bhattacharyya et al., 2011; Mohan et al., 2006; Xie et al., 2019).
 188 The adsorption isotherm is a curve that reveals the equilibrium relationship between the
 189 concentration of solute molecules in two phases at a particular temperature when the adsorption
 190 process is complete at the interface of these phases. An accurate mathematical model for the
 191 adsorption isotherm can simplify the properties of adsorbents and the adsorption capacity of Cr
 192 (VI), as well as help in the removal mechanism between adsorbents and Cr (VI). Brunauer–
 193 Emmett–Teller adsorption isotherm, Dubinin–Radushkevich equation, Langmuir, Freundlich,
 194 Temkin, Redlich–Peterson, Kelvin equation, Toth and Sip model have been used as adsorption
 195 isotherm models (Villarroel Rocha et al., 2011; Saadi et al., 2015; Tran et al., 2017). The study of
 196 adsorption thermodynamics allows for an understanding of the extent and driving factors behind

197 the removal of Cr (VI), as well as an in-depth analysis of the diverse influences of various factors
198 on the adsorption process. The enthalpy change (ΔH_0) is determined using the Cladis–Clapeyron
199 equation, while the Gibbs free energy change (ΔG_0) and the entropy change (ΔS_0) of the adsorption
200 process are computed through pertinent statistical thermodynamic formulas and the Gibbs–
201 Helmholtz equation once adsorption reaches equilibrium.

202 Hassan and Garrison (1996) collected three soil samples (glacially derived soil, loose soil
203 sample, and clay soil) to identify the influencing, partitioning, and interconversion process.
204 Adsorption isotherms measured have been used to calculate K_d for each of the soil samples for Cr
205 (III) and Cr (VI). The equilibrium constant (K_d) values, obtained under natural soil-groundwater
206 systems with a consistent equilibration time, exhibit significantly lower magnitudes for Cr (VI),
207 falling within the range of <1 to ~50 l/kg across the three soils examined. In contrast, for Cr (III),
208 the K_d values range from approximately 850 to 5,600 l/kg. Notably, the total organic carbon (TOC)
209 concentration in the soil exerts a significant influence on the K_d values for both Cr (III) and Cr
210 (VI) as the highest value of K_d found in the clay soil which has the maximum TOC contain.

211 McBride et al. (1997) found if the K_d value will be high for Cr, with a very high K_d value,
212 then it will be the least mobile element. This observation reinforced their initial choice of using Cr
213 as the reference element in soil. This reference element, with its high K_d value, served as an
214 indicator for assessing the leaching potential of other elements in the soil. Moreover, Gao et al.
215 found that Cr as CrO_4^{2-} instead of Cr^{3+} was the last element in the sequence of adsorption with
216 the lowest K_d value. Ramnani and Sabharwal (2005) found that the adsorption of Cr (VI) was
217 maximum at pH 3 on cross-linked chitosan. Equilibrium adsorption studies done using Langmuir,
218 Freundlich, and Dubinin–Radushkevich (D–R) models to analyse results and evaluate model
219 parameters.

220 Castr-Rodriguez et al. (2015) analysed the adsorption capacity of the soils through
221 adsorption isotherms, enabling the determination of soil particle charge and the Gibbs free energy.
222 They collected a total of 3 soil samples for analysis. The mean concentration of total Cr was 43.5
223 mg/kg, whereas the average concentration of Cr (VI) was 4.37 mg/kg. Adsorption isotherms were
224 used, and the linear, Langmuir, and Freundlich models were employed for analysis. The Freundlich
225 model exhibited the best fit for two soil samples, while the linear model provided the best fit for
226 another soil. For a sample which contains fine-grained sand, sandy silt the adsorption level was
227 lowest ($K_d = 0.0049 \text{ ml/g}$) whereas soil samples which contain a high clay the highest value (K_d
228 = 0.7 ml/g) was obtained. The average adsorption of Cr (VI) was 0.34 ml/g.

229 Zhang et al. (2019) studied adsorption/reduction of Cr (VI) by a black soil. This study
230 focused on examining the adsorption and reduction kinetics of Cr (VI) using a representative black
231 soil. The research explores different initial Cr (VI) concentrations (ranging from 40 to 400 mg/l),
232 and pH varies between 3.5 to 7.0. At pH 5.7, a nearly linear partition relationship was observed
233 between the adsorbed Cr (VI) content and the remaining Cr (VI) concentration in the solution.

234 This relationship showed a good fit curve with both the linear model ($R^2 = 0.98$) and the Freundlich
235 model ($R^2 = 0.99$). As the pH decreased, the partition behaviour exhibited a non-linear pattern, and
236 the Freundlich model provided a better description of non-linearity.

237 He et al. (2022) simulated the migration as well as transformation of Cr (VI) from a
238 contaminated site by ferroalloy factory. The experimental data indicates that the Cr (VI) adsorption
239 by silty clay is well-fitted by the Langmuir isotherm model. The maximum adsorption capacity of
240 silty clay for Cr (VI) is 466.6 mg/kg. The hydraulic conductivity of silty clay, when subjected to
241 infiltration by distilled water and a 160 mg/l Cr (VI) solution, ranged between 6.5×10^{-7} and
242 6.7×10^{-7} cm/s. The hydrodynamic dispersion coefficient (D) for silty clay was determined to be
243 1.4×10^{-4} m²/d. The retardation factor (R) was found to be in the range of 4.2 to 10.

244 Cao et al. (2021) investigated the adsorption mechanism of Cr (VI) in sediments with
245 different properties to identify the controlling factors involved in the process. The research
246 involved the evaluation of Cr (VI) adsorption on six natural unconsolidated sediments, including
247 two loamy sands, two sandy loams, one loam, and one silty loam clay which were collected from
248 a fluvial aquifer located in Qiqihar, China. The study evaluated adsorption kinetics with Elovich,
249 pseudo first- and second-order models and equilibrium properties using Henry, Freundlich, and
250 Langmuir isotherms. Higher solution pH and solid-to-solution ratios reduced Cr (VI) adsorption.
251 Authors reported that sediment properties, especially grain size and clay mineral content, were key
252 factors. Smaller grain sizes and higher clay mineral content have more adsorption capacity and
253 slower adsorption rates.

254 Ye et al. (2019) collected 6 soil samples from chromium-containing slag from Xiangxiang,
255 China and performed batch and column experiments for adsorption of Cr (VI) in soil. To
256 investigate the absorption mechanism kinetic models namely pseudo-first order and pseudo-
257 second order have been tested and the results shows that the pseudo- second order kinetic model
258 fits very well for adsorption of Cr (VI).

259

260 **Table 3: Summary of studies performed to investigate Cr adsorption in soil.**

Citation	Objectives	Study Area	Experimental Conditions and Methods	Adsorption Coff value	Key Observation
Ye et al. (2019)	Investigate soil Cr (VI) adsorption, develop adsorption models, explore Cr (VI) transport behavior, and predict Cr(VI) leaching from chromium-containing	Xiangxiang city (112°28'42.91"N, 27°44'50.96"E)	Scale: Batch scale and column experiment (40 cm hight and 7 cm diameter)	$K_d = 1.17$ cm ³ /g $R_d = 10.6$	The concentration levels of Cr (VI) in the soil-leaching solution attained 905 mg/L.

	slag in soil via mathematical modeling.		Models: Kinetic models of adsorption, Isotherm models of adsorption, Theoretical modelling of transport		The pseudo-second-order kinetic model fits very well with the dynamical adsorption behavior of Cr (VI). Langmuir isotherm better fit the equilibrium data for Cr(VI) adsorption.
Khan et al. (2010)	Develop models to understand hexavalent chromium sorption and transport in soil media.		Scale: Column experiment Analytical Method: UV spectrophotometer	$K_d = 0.0115 \text{ cm}^3/\text{g}$ $Rd = 2$	The initial concentration of Cr (VI) in the solution remarkably influenced the equilibrium Cr (VI) uptake. The periodic movement of the inorganic contaminant concentration front ultimately reaching quite low concentration at the end of the cycles.
Wu et al. (2022)	To develop a mathematical model for quantitatively estimating and effectively predicting Cr pollution in the "slag-soil-groundwater" system by integrating Cr (VI) migration and leaching transport data.	Xiangxiang City, China	Scale: Column experiment (height 81 cm)	$K_d = 0.0542 \text{ cm}^3/\text{g}$ $Rd = 1.17$	The total amount of leached Cr (VI) and the concentration of Cr (VI) slag present a rising trend. Measured and simulated values exhibited correspondence, with a maximum

					RE of 31%, a minimum RE of 12%, a ME of 0.221, and an RMSE of 0.430.
Ektre et al. (2007)	to comprehend the migration of Cr in diverse soil types and to create models for this purpose.	Guangxi Province, China	Scale: Batch scale Analytical Method: UV spectrophotometer Models: Kinetic models of adsorption, Langmuir isotherm	$K_d = 0.234 \text{ cm}^2/\text{g}$ $R_d = 2.21$	the results of the adsorption experiment indicates that Langmuir isotherm better fit the equilibrium data for Cr (VI) adsorption
Yan et al. (2023)	To understand Cr migration and its interaction with different soil types, and the associated long-term risks in studied sites.	Shijiazhuang (38°02'33"N and 114°30'51"E), Zhuzhou (27°49'40"N and 113°08'02"E) and Guangzhou (23°07'45"N and 113°15'22"E), China	Scale: Column experiment (1.2 cm diameter and 8 cm height) Analytical Method: UV spectrophotometer	$K_d = 0.84 \text{ cm}^2/\text{g}$ $R_d = 1.43$	The soil of Zhuzhou had the maximum adsorption capacity for Cr (VI) (0.225mol/kg). Rainfall intensity impacted Cr (VI): heavy rain increased vertical risk; light rain aided reduction.
Arthur et al. (2017)	Examine DNAN fate in soils, quantify reversible adsorption, assess mass loss, and analyze DNAN-soil property relationships.	Entisols, Inceptisols, Mollisols, Aridisols, and Ultisols in USA	Scale: Batch adsorption Analytical method: high performance liquid chromatograph (HPLC) Models: Freundlich and linear isotherms	$K_d = 0.6 \text{ to } 6.3 \text{ L/kg}$ $R_d = 3.5 \text{ to } 27.5$	The equilibrium adsorption data were fitted to Freundlich and linear isotherms with the Freundlich isotherm resulting in slightly better fit on average.

Jardin e et al (2011)	To investigate the fate and transport of Cr (III) and Cr (VI) in three different soil types	Oak Ridge National Laboratory (ORNL) eastern Tennessee; Savannah River National Laboratory (SRL) South Carolina and Hanford facility Washington	Scale: Batch scale and column experiment Model: Freundlich isotherms	$K_d = 0.2 \text{ cm}^3/\text{g}$ (Hanford soil), 14 cm^3/g (ORNL) and 5 cm^3/g (SRL)	The transportation of contaminants in the environment is determined by whether the sorption processes are linear or nonlinear, and whether they are kinetically controlled or in equilibrium with the soil solution. Soil chemical and mineralogical properties exert significant control on time-dependent Cr–solid phase interactions, influencing reactive processes.
Cao et al. (2021)	to evaluate the adsorption and transport of Cr (VI) on several natural sediments	West of Songnen Plain, China	Scale: Batch scale and column experiment Models: Kinetic models and Equilibrium adsorption isotherms	$K_d = 0.897 \text{ to } 6.430 \text{ L/kg}$ $R_d = 2.86 \text{ to } 71$	The order of adsorption capacity in different soil was loam, silty loam clay, sandy loam, and loamy sand, respectively. The adsorption of Cr (VI) decreases with increase of

					the pH and solid/solution ratio.
Liu et al. (2019)	To explore the adsorption characteristics of Cr (VI) in different aquifers and seeking insights into the underlying mechanisms governing Cr (VI) adsorption.	Tianjin Tongsheng Chemical Factory, Tianjin, China (39°14'21.4"N and 117°06'28.3"E).	Scale: Batch scale and column scale Models: Langmuir model and Freundlich mode	K _d = 0.052 L/kg to 2.230 L/kg	The adsorption capacities of Cr (VI) on three aquifers followed the order as clay > silty clay > silt. Cr (VI) is not easily adsorbed by the aquifer mediums but transports with groundwater.

Hellerich and Nikolaidis (2005)	To define and quantify the capacity and kinetics of the Cr (VI) attenuation processes in the environment	Connecticut US state	Scale: Batch scale,	$K_d = 1.1 \text{ mL/g}$ to 4467 mL/g	Chromium attenuation took place quickly in wetlands soils. The elimination of aqueous phase Cr (VI) from aqueous, soil slurries was obtained in moments at lower pH's.
Yolcu bal and Akyol (2007)	to investigate Cr (VI) sorption and transport in a high alkaline and calcareous soil under varied saturated flow conditions	Karstic Travertine Plateau, Antalya Turkish Mediterranean	Scale: Batch Scale, Column experiment Models: deterministic equilibrium and physical nonequilibrium (two region)	$K_d = 0.097 \text{ L/kg}$ (from saturated column experiment) and 0.53 L/kg (from batch experiment experiment)	Elevated K_d 's (more than 250 mL/g) noted at environmentally relevant pHs of 6-6.5 suggest that Cr(VI) is being reduced to Cr(III), followed by precipitation or complexation with SOM functional groups. At both saturated and unsaturated flow conditions, Cr (VI) transport was slightly delayed compared to non-reactive tracers. Column experiments with low retardation

			solute transport models	$R_d = 1.34$ and 1.07	factors and K_d values indicate that Cr (VI) species are relatively mobile and bioavailable in highly calcareous soil conditions.
262	Khan et al. (2010)	To investigate the adsorption of Cr(VI) from aqueous solution by soil in batch and continuous mode of operations	Scale: Batch Analytical method: UV-vis spectrophotometer Models: Freundlich isotherm, Langmuir isotherm	$K_d = 0.0115$ to 0.0087 mg/g	Simulated soil redemptions cycles show periodic chromium concentration fluctuations, reaching low levels by the cycle's end. The concentration of contaminants reaching the ground water table becomes quite low.

262

263 **3.2 Precipitation**

264 The precipitation of Cr is a critical aspect of Cr chemistry, involving the controlled
 265 conversion of soluble Cr in aqueous solutions into solid, typically crystalline, phases, with
 266 significant implications for environmental remediation applications. Chromium can precipitate in
 267 response to changing environmental conditions in the soil-groundwater system (Bodek et
 268 al., 1988). The most used method for removing Cr (VI) involves its reduction to Cr (III)) and the
 269 subsequent precipitation of Cr (III) as chromium hydroxide, a process often achieved using sulfur
 270 compounds like sulfur dioxide (SO_2) and sodium bisulfite ($NaHSO_3$) (Barrera-Diaz et al., 2012).
 271 The parameters influencing the precipitation process include the precipitation agent, pH level,
 272 agitation intensity or speed, and the presence of complexing agents (Abdulla et al., 2010).
 273 Traditional chemical precipitation methods involve the use of sulfide precipitants and hydroxide
 274 precipitants (USEPA 2000). Chromium is often precipitated by compounds like calcium hydroxide

275 [Ca (OH)₂], sodium hydroxide (NaOH), magnesium oxide (MgO), and calcium magnesium
276 carbonate [Ca-Mg (CO₃)₂] (Hintermeyer et al., 2008).

277 In their study, experimental data detailing the kinetics of chromium precipitation from
278 chromium (III) acetate solutions was presented by Jou et al. (2000). Precipitation occurs by the
279 rise in solution pH resulting from fluid-rock interactions, predominantly in carbonate rocks. This
280 study was carried out on pH ranging between 7 to 10 and the temperature varies between 25 to 45
281 C. Elevated temperature, salinity, and higher divalent ion concentration increase precipitation rates
282 and can make the sub-surface placement of chromium acetate-polyacrylamide gels in carbonate
283 matrix rock less effective. Tingting (2021) studied the effect of temperature on precipitation of Cr.
284 The precipitation of Cr has been studied on temperature 20, 40 and 60 C. The finding of the study
285 shows that the maximum precipitation rate was 60 C whereas the minimum precipitation occurs at
286 20 C which concluded that the precipitation of Cr increased with increase in temperature. Dutrizac
287 and Chen studied Cr (III) precipitation in jarosite compounds. Formation of the Cr (III) analogue
288 of potassium jarosite at 98°C failed, resulting in an amorphous phase with low K content. These
289 analogues were not formed at temperatures below 100°C. However, successful Cr (III) substitution
290 for Fe (III) in potassium jarosite and sodium jarosite occurred. The Cr (III) analogue of potassium
291 jarosite (KCr₃(SO₄)₂(OH)₆) was precipitated at 200°C. To comprehend the fate and transport of
292 Cr (III), it is essential to differentiate between homogeneous and heterogeneous precipitation,
293 quantify the volume of precipitates, and elucidate the mechanisms responsible for Cr(III)
294 sequestration within precipitates (Dai et al., 2016). Aqueous Cr (III)/Fe (III) ratios were found to
295 determine the homogeneous and heterogeneous precipitation rates of (Fe_x, Cr_{1-x}) (OH)₃ and affect
296 the mechanisms of Cr (III) sequestration in the (Fe_x, Cr_{1-x}) (OH)₃ precipitates.

297 **3.3 Dissolution**

298 The Cr dissolution in soil-groundwater is a process governed by various physicochemical
299 factors such as pH, temperature, redox and involves the interplay of different oxidation states of
300 chromium. Low pH and reducing conditions favour the dissolution of Cr (III) from solid phases,
301 while Cr (VI) solubility is higher under alkaline conditions (Richard and Bourg, 1991). Organic
302 matter and complexing ligands in groundwater can further affect the solubility of both Cr (III) and
303 Cr (VI). Cr (VI) is normally soluble and rarely forms precipitates; nevertheless, it can become
304 integrated into specific solid phases by substituting oxyanions, particularly due to sulphates and
305 carbonates (Baron and Palmer, 1996). Eary and Zachara (1989) highlighted the importance of
306 thermochemical data on Cr (III) and Cr (VI) solid phases with fast dissolution kinetics and low
307 solubility. This data is crucial for comprehending the nature of Cr in natural environments and
308 managing Cr contamination in soil-groundwater systems. Pettine and Capri (2005) analysed the
309 kinetic and thermodynamic characteristics of reactions involving the dissolution of Cr (VI)
310 indicate that at higher pH levels (>10), there is a lower risk of Cr (VI) reduction. The faster reaction
311 kinetics of oxygen divert the reductants away from Cr (VI), reducing the likelihood of its
312 conversion to more toxic Cr (III) forms. Shi et al. (2021) investigated the photo-reductive

313 dissolution of Cr (VI) in the presence of tartaric acid, exploring various influencing factors, such
314 as pH, TA concentrations, in the presence of coexisting ions including Al^{3+} , Ca^{2+} , K^+ , and CO_3^{2-} .
315 The results showed that the combination of TA and simulated solar radiation significantly
316 enhanced the release of TFe from the solid phase containing Cr (VI) in the study (SCHeCr (VI)).
317 The produced Cr (III) either adheres to the mineral surface or combines with dissolved Fe (III) to
318 form FeCr(OH)_6 precipitates. Lower pH conditions facilitate the release of both TFe and total Cr,
319 while potassium (K^+) and calcium (Ca^{2+}) ions had negligible effects on their liberation.
320 Chrysochoou et al. (2016) found that Cr emitting into groundwater involves the dissolution of Cr
321 (III) from minerals, followed by its sorption onto high-valence Mn oxides and oxidation to mobile
322 Cr (VI) under alkaline pH.

323 **3.4 Leachability**

324 Leachability of contaminants is the process by which pollutants migrate through soil
325 downward, assisted by water seeping through. Typically, more precipitation increases the
326 likelihood of contaminants leaching, which is concerning because these contaminants could seep
327 into the soil-groundwater system (Table 4). The chromium compounds (Cr^{3+} and Cr^{6+}) that leach
328 through groundwater are dependent upon the effluent's presence, gradient flow direction, and
329 beginning concentration. The main forms of Cr (VI) are bichromate (HCrO_4^-) and chromate (CrO_4^{2-}),
330 which are highly soluble in most environmental pH ranges of 6.5 to 7.5. Even at quantities
331 lower than 50 $\mu\text{g/kg}$, CrO_4^{2-} is poisonous to a wide range of plants and animals, oxidant, mutagenic,
332 and carcinogenic. At pH levels above circumneutral, Cr (III) is comparatively non-toxic and
333 sparingly soluble in hydroxide, forming mixed hydroxide precipitates with Fe and Cr. Because of
334 its low solubility, it is only weakly mobile in groundwater, and the precipitated Cr(OH)_3 interacts
335 slowly with molecular oxygen (Tripathi and Chaurasia, 2020).

336 The chemical speciation of Cr is important for determining the leachability linked with Cr
337 contaminated soil (Hu et al., 2016). The stability and leachability of Cr (VI) and Cr are important
338 elements impacting the effectiveness of stabilized Cr (VI)-contaminated soil. (Jacobs et al. 2005)
339 found that redox potential, soil pH, and solution pH had a significant impact on Cr (III)
340 leachability. As amorphous Cr(OH)_3 degraded into soluble chromium hydroxide cations at low
341 pH (below 6), the solubility of Cr increased. The lowest leachability occurred between pH 6 and
342 11. Around pH 8.0, insoluble Cr(OH)_3 was seen as the circumstances were reducing.
343 Kanchinadham et al. (2013) conducted a leachability study of Cr and Cr (VI) for COPR and soil
344 samples using acetic acid and water as leaching medium. Authors found that, Cr (III) only leached
345 in acetic acid because it was insoluble at $\text{pH} > 6$, Cr (VI) leached out in both media. Zhang et al.
346 (2021) studied leachability of Cr in different grain size of contaminated soil. The contaminated
347 soil was divided into bulk soil, coarse soil, medium sand, fine sand and silt-clay. The proportions
348 of fine sand (43.25%) and silt clay (43.26%) exhibited greater leachability of chromium compared
349 to bulk soil (32.04%). Furthermore, compared to bulk soil (68.77%), the leachability of Cr (VI)

350 was higher in silt-clay (85.72%) and fine sand (75.19%). Hence, as the particle size decreased, the
351 concentrations of Cr and Cr(VI) in the leachate increased.

352 **Table 4: Summary of studies performed to investigate Leachability Index**

Citation	Objectives	Study Area	Experimental Conditions and Methods	Leachability Index	Key Observation
Kameswari et al. (2015)	to develop a treatment procedure for the solidification and stabilization (S/S) of CCS caused by Cr (VI) leaching from COPR	Ranipat Tamil Nadu	Leachability test, Compressive strength test	LI = 7.99 – 9.01	Cr (VI) and total chromium contents in the contaminated soil were as high as 18,770 mg/kg and 44,615 mg/kg, respectively. LI was mostly between 8 and 9, showing that FA is a good treatment for CCS disposal into secured landfills.
Zhang and Lin (2020)	To investigate the influence of redox conditions for Cr release from Cr contaminated soil	Shanghai, China	Semi-dynamic leaching test, TCLP test, Sequential chemical extraction test	LI = 8.52 – 10.27	Maximum amount of Cr leached on strong oxidation condition ($E_h = 400$ mV). Cr leaching was strongly dependent on E_h .

Sophia and Swaminathan (2005)	to use Portland cement and fly ash as hydraulic binders for solidifying electroplating waste sludge with high chromium and metal concentrations, with various tests to assess post-solidification leaching potential.	Tamil Nadu	TCLP Test, Spectroscopy,	LI = 15.2 – 16.9	TCLP concentration of Cr in the leachate was lesser than the USEPA regulatory limit. Diffusion controls the leaching mechanism of the waste.
Panda et al. (2022)	To determine the chromium immobilization and resultant leaching from unbonded and concrete bonded ferrochromium slag.	Kalinga Nagar, India	XRF, XRD, SEM, TCLP	LI = 14.6 – 15.2	As per the comprehensive leaching investigation, the release of Cr from unbound slag frequently exceeds the Indian regulatory discharge norms for Cr (VI).

Liu et al. (2018)	to investigate the leachability of PTMs Pb, Zn, Cd, Co, Cr, Cu, and Ni from zinc smelting slag materials-amended acidic soils under acid deposition conditions, and to simulate the probable chemical species of PTMs in the leachate.	Guizhou, China	Column leaching experiment, Leaching Index	LI = 7.1 - 11	The leaching process was affected by the content of the leachate as well as the geochemical features of PTM-associated minerals. The primary PTM speciation in leachate was free metallic ions, while metallic sulphate complex factored in as SSM amendment ratios increased in soils.
Taghi pour and Jalali (2023)	To investigate the leaching behaviour of Cr, Cu, Ni and Zn in four different industrial solid waste.	Hamadan, Iran	Column leaching experiment	LI = 0 – 62.52	The highest cumulative amount of Cr leaching was made from the Ceramic factory waste and Leather factory waste.

Toma sevic et al. (2014)	to evaluate stabilization treatments in landfill-like circumstances, identify As, Pb, and Zn leaching mechanisms, estimate environmental damage through various leaching procedures, and evaluate the efficacy of bentonite- and kaolinite-nZVI for immobilization.	Nadela river basin, Vojvodina, Serbia	Microwave-assisted sequential extraction, ANS method, TCLP test, Synthetic precipitation leaching procedure (SPLP test), Waste extraction test (WET test)	LI = 7.46 – 10.67	Making LI values, diffusion coefficients, and single leach test results into account, the applied treatments demonstrate efficacy in immobilizing variously distributed metals, providing an economic justification for implementing costly remediation procedures, especially for materials containing a mixture of pollutants.
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Wang et al. (2022)	to validate the environmental safety of Cr in established material using several static batch leaching experiments, and to assess the release mechanisms of Cr in solidified material using long-term mass transport-based leaching tests and dynamic droplet leaching experiments	Beijing, China	Leaching test	LI = 11.52 – 12.08	The mechanism of Cr leached from the solidified material was mainly diffusion. Because the mean leaching index values were larger than 9, the treatment of contaminants is regarded effective, and the waste-containing material can be used for various building purposes.
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353

354 3.5 Transport of Chromium

355 The chromium, which primarily occurs as Cr (III) and Cr (VI), exhibit different transport
 356 behaviours. Cr (III) has a propensity to adhere firmly to soil particles and create insoluble
 357 precipitates, which reduces its mobility. The pH, organic matter content, and mineral composition
 358 of the soil are important factors that affect Cr (III) transport. On the other hand, Cr (VI), which is
 359 more soluble, is vulnerable to redox changes, and the pH and prevailing redox conditions have an
 360 impact on its mobility. Advection, dispersion, sorption, and degradation are some of the physical
 361 and biological mechanisms that cause chromium to migrate through soil. The two primary
 362 mechanisms that allow Cr (VI) to transport from the subsurface to the ground surface are capillary
 363 rise and evaporative enrichment, and evaporative enrichment (Henry et al., 2007). Contaminants
 364 may rise to the surface through capillary action when the capillary rise is higher than the distance
 365 between the water table and the surface (Henry et al., 2007; Hillel, 1998).

366 3.5.1 Advection

367 Advection is the movement of dissolved Cr with soil-groundwater systems. The amount of
368 Cr being transported is a function of its concentration in the groundwater and the quantity of
369 groundwater flowing. Advection, as it relates to chromium transport, is the convective flow of
370 chromium species in a soil-groundwater system. Advection of chromium, for example, occurs in
371 the soil-water system when chromium (III) and chromium (VI) species are displaced by the
372 moving water phase. The behaviour of chromium during advection is influenced by its speciation,
373 with hexavalent chromium exhibiting higher mobility due to its lower propensity for adsorption to
374 soil particles compared to trivalent chromium. Chen et al. (2021) found that advection is the
375 dominant transport mode that highly controls the Cr transport. Soils with lower permeability,
376 including montmorillonite and kaolinite, show significant delays in the transfer of Cr. High
377 montmorillonite content causes the slowest Cr movement and the lowest peak concentration in
378 simulations, as it does in natural clay and montmorillonite soils.

379 **3.5.2 Diffusion**

380 Diffusion is the movement of Cr in water from an area of higher concentration to an area
381 of lower concentration (Agah et al., 2017). Even in the absence of fluid movement, diffusion will
382 take place as long as there is a concentration gradient, which means that a contaminant may diffuse
383 outward from the site of injection into a porous medium. If each adjacent stratum has the necessary
384 porosity, diffusion can also occur when a contaminant's concentration is higher in one stratum than
385 in another. The rate of diffusion (Q_d) at rest is given by Fick's Law:

386
$$q_d = -D_e \frac{\partial C}{\partial x}$$

387 Where D_e (m^2s^{-1}) is the coefficient of diffusion and C (kg m^{-3}) is solute concentration in
388 soil-groundwater system. The minus sign shows the direction of diffusion is opposite to the
389 direction of increasing concentration. Diffusion coefficient in soil-groundwater system is
390 determined by mathematical analysis as a function of position or time.

391 **3.5.3 Dispersion**

392 Dispersion in the soil-groundwater system refers to the process by which solutes put into
393 the soil are disseminated or distributed in the groundwater as it moves through the subsurface.
394 Differences in the flow velocities due to the different pore size and shape (area and volume of the
395 solute) cause solute transport at different rates and thus lead to dispersion or mixing of an incoming
396 solution. Furthermore, the dispersion may occur in soil-groundwater system due to the
397 development of a velocity profile within an individual pore such as the highest velocity occurs in
398 centre and no flow or negligible flow at the walls, different mean velocities of different size of
399 pore, the mean water flow direction being different from the actual streamlines in the soil-
400 groundwater system and solute particles diverging or converging from the same pore.

401 The solute flux is given by an equation below.

402

$$q_h = -D_h \frac{\partial C}{\partial x}$$

403 Where q_h (m/s) is the solute dispersive flux D_h dispersion coefficient ($m^2 s^{-1}$), C ($kg m^{-3}$) is
404 solute concentration in soil-groundwater system (Table 5).

405 **Table 5: Summary of studies performed to investigate Dispersion Coefficient**

Citation	Objectives	Study Area	Experimental Conditions and Methods	Dispersion Coff value	Key Observation
Yan et al. (2023)	to study the fate and transport of Cr in soil and assess the risks associated with longterm vertical transport of Cr	Shijiazhuang, Zhuzhou and Guangzhou, (China)	Scale: Column experiment Model: Saturated water flow model, Solute transport model,	$D = 7.84 \text{ cm}^2/\text{h}$	the horizontal transport of pollutants caused by groundwater will also lead to the diffusion of heavy metal.
Trento and Alvar ez (2011)	to build and utilize an Eulerian numerical model to simulate non-steady chromium transport in water, bed sediments, and fine sediments suspended in a stream		Scale: Column Models: Mathematical model, Numerical model	$D = 10.6 \text{ m}^2/\text{cm}$	The Salado River is fully affected by Chromium. An accurate approximation of chromium concentrations in water and bed sediments.

Khan et al. (2010)	To model the transport of Cr (VI) through soil and estimated the time for reaching the contaminant in layers of aquifers		Scale: Column experiment Analytical Method: UV spectrophotometer Models: Adsorption isotherm, Freundlich isotherm, Langmuir adsorption isotherm	$D = 0.00475 \text{ cm}^2/\text{h}$, $0.00732 \text{ cm}^2/\text{h}$ and 0.00768 at 1.2 ml/min , 1.0 ml/min and 0.8 ml/min flow rate respectively.	The initial concentration of Cr (VI) in the solution remarkably influenced the equilibrium Cr (VI) uptake. The periodic movement of the inorganic contaminant concentration front ultimately reaching quite low concentration at the end of the cycles.
Selim et al. (1989)	To investigate the mobility of Cr (VI) in six soils with different properties and develop a transport model for the Cr (VI) interactions and movement in soil		Models: Kinetic model, Convective-dispersive transport equation,	$D = 0.0001296 \text{ to } 0.0004464 \text{ m}^2/\text{d}$	The model provided inadequate ETC predictions for the highly reactive soils. When model parameters were produced using a nonlinear least square (best-fit) parameter optimization approach, the model could provide a good description of Cr BTCs for all soils.

Boupha et al. (2004)	To develop a software library and user interface that provide solutions to contaminant transport equations to invert Laplace transforms		Models: Laplace transforms, The De Hoog algorithm	$D = 0.2 \text{ m}^2/\text{day}$	The purpose of the program is to readily provide solutions to encourage a better understanding of factors affecting the fate and transport of groundwater contaminants.
Yolcu bal and Akyol (2007)	To determine the sorption & transport of Cr (VI) in a high alkaline and calcareous soil.	Antalya, Turkish Mediterranean coast	Scale: Batch experiment and Column experiment	$D = 44.7 \text{ cm}^2/\text{hr}$ (saturated experiment) and $0.3 \text{ cm}^2/\text{hr}$ in unsaturated condition	The outcomes of the column experiments revealed that Cr (VI) transport was marginally delayed compared to nonreactive tracer at both saturated and The high mobility of Cr (VI) enhances the rate of pollutant distribution and the potential for groundwater contamination flow conditions.

	Al-Rahman et al. (2006)	To study the different transport mechanisms and soil interaction on the transport of Cr (VI) in groundwater.	Liwa	Scale: Batch experiment, Column experiment Model: CXTFIT2	D = 0.93 cm ² /hr, 2.72 cm ² /hr and 2.75 cm ² /hr	The capacity of Liwa sand to absorb Cr (VI) is minimal, whereas its ability to remove Cr (III) from the solution is significant. The dispersivity of columns packed with Liwa sand averages 0.4 cm.
406	Qiu et al. (2012)	To develop a 1-D mathematical model basing on advection–diffusion equation, to describe the fate of contaminant in the river.		Scale: Laboratory scale Model: Transport-dynamic model	D = 0.00096 to 0.0088 m ² /min	The fate of Cr (VI) in the river was well described, by using mathematical model. Almost no toxicity was detected in the treated solution even after 36 days.

406

407 **4. Role of Aquifer Materials & Dynamics in Fate and Transport**

408 There are many environmental factors that influence the fate and transport of chromium in
 409 groundwater and aquifers. Type of rocks, precipitation, moisture content, hydraulic conductivity,
 410 different soil properties and organic matter content and water table fluctuations are the main
 411 components which affect the fate and transport in the soil-groundwater system. Particularly in
 412 India, the groundwater characteristic is very complicated due to different climate conditions,
 413 different geology, litho-logical characteristics, and different hydro-chemical characteristics. The
 414 hydrogeological formation of India is divided into the 6 different zone (Kulkarni et al., 2015).

415 **4.1 Heterogeneity of surface material**

416 In a heterogeneous system, Cr will undergo various transformations that can alter the fate
417 and transport as well as toxicity. Understanding various factors which affect the environmental
418 dynamics of Cr is essential to know the interactions between the Cr and heterogenous soil-
419 groundwater system. Surface sediments are a heterogeneous and complex material for fate and
420 transport of Cr (Table 5). Dynamics of Cr is important for understanding as the matrix is composed
421 of pore fluids, inorganic solid particles, organic matter, and living organisms in which various
422 transfers between phases can occur. Although the sediment compartment can be heterogeneous,
423 spreading punctual characterizations as well as monitoring a specific localization of Cr as a
424 function of time can be difficult. Second, large redox gradients can occur in sediments, particularly
425 in surface sediments where bacterial action mineralizes biodegradable organic materials, including
426 the consumption of dissolved oxygen and other oxidants (Froelich et al., 1979).

427 Soils and sediments in the vadose zone frequently do not contain enough naturally
428 occurring inorganic reductants to influence the redox state of Cr (VI). Aerobic soils and sediments
429 usually contain significant amounts of natural organic matter, which may accelerate the reduction
430 of Cr (VI) to the less soluble, less mobile Cr (III) species (Jardine et al., 1999). Xu et al. (2020)
431 revealed that the behaviour of metal (Cr) in heterogeneous systems is different from the behaviour
432 in homogeneous systems especially in the soil media. They studied the effect of soil pH on the
433 transport, fractionation, and oxidation of Cr (III). The results showed that alkaline soil retards the
434 transport of Cr (III) whereas in acidic soil transport of Cr (III) easily occurs. Moreover, the high
435 retention of Cr (III) was mainly attributed to the precipitate at pH 8.0 soil. Yan et al. (2023) studied
436 migration and transformation of Cr in the heterogeneous soil by collecting soil from various depths
437 in Shijiazhuang, Zhuzhou, and Guangzhou in China. They observed the Ch has greater mobility in
438 the soil collected from Shijiazhuang which may be an alkaline soil. Cr (VI) was faster transport
439 in the upper soil of Guangzhou while subsoil has a strong retarding ability owing to high Fe and
440 TOC content. Kantar et al. (2008) conducted a study to determine the effects of exopolymeric
441 substances (EPS) on Cr (VI) transportation and stabilization in heterogeneous soil systems. The
442 presence of organic matter increases Cr (VI) uptake by soil surfaces under acidic to slightly
443 alkaline pH conditions, depending on the concentration of organic ligands and pH. Fendorf et al.
444 (2000) also concluded that due to the heterogeneous and complex nature of soils and surface water,
445 Cr fate determination is a most difficult task.

446 4. 2 Effect of Groundwater Table

447 The groundwater table and its associated capillary fringe function as a transition zone
448 between unsaturated and saturated zones in shallow subsurface situations. Natural processes such
449 as seasonal rain and being dried, groundwater-surface water interaction, and tidal processes cause
450 groundwater table fluctuations (Duan et al., 2015). Rainfall infiltration has a great effect on the
451 groundwater table fluctuation (GTF) which linearly depends on the effective accumulated rainfall
452 amount (Jan et al. 2007). Human activities such as groundwater extraction for drinking and
453 irrigation, water conservation initiatives, and artificial groundwater replenishment can all

454 contribute to the variation (Deng et al., 2016). The fluctuation in groundwater table is not only the
455 process by which water exchanges between unsaturated and saturated aquifers, but it is also the
456 main interface via which contaminants migrate from the surface into saturated aquifers (Sreedevi,
457 2002). The groundwater table variation affects the redox environment and leads to the
458 hydrogeochemical components change. The change in the water content of the unsaturated zone
459 during fluctuation caused by rainfall affects the redox environment. Using a two-dimensional sand
460 tank (Gupta et al., 2019) and three-dimensional tank (Gupta and Yadav 2020), found the fate and
461 transit of toluene in the subsurface region under dynamic groundwater table conditions.

462 Wei et al. (2021) investigated Cr (VI) migration and transformation in the unsaturated zone
463 by water table fluctuation. Infiltration and groundwater drainage were the two main elements
464 which influence water content, and water level in the unsaturated zone. Water table fluctuations
465 caused Cr (VI) to migrate downwards and reduce. Wei et al. (2023) provided a colloid-facilitated
466 Cr transport model taking variable density flow during groundwater table fluctuations into
467 freshwater and seawater. The study revealed that the fluctuation may affect the migration and
468 transportation of Cr contaminants.

469 4.3 Hydrology of Sites

470 Climate change is predicted to affect precipitation, evapotranspiration, and runoff rates,
471 and the dynamics of hydrogeological processes, as well as the quantity and quality of water in
472 aquifers (Narula and Gosain, 2013 and Ercan, 2020). The groundwater-soil system is very
473 complicated, and groundwater table always rise with precipitation and river feeding which lead to
474 the changing of soil properties, and then change the migration and transformation of Cr. The
475 chemistry of the subsurface environment, which is governed by the hydrology and composition of
476 the aquifer substrates, governs the discharge of geogenic contaminants into groundwater (Peel et
477 al., 2022). Cr tend to travel vertically to deeper layers rather than being retained in topsoil due to
478 solid residue percolation, and surface runoff from rainfall. The thickness of the mixing layer
479 determines the instantaneous mixing of soil water and surface runoff, as well as the complete or
480 incomplete adaptation of precipitation in a shallow layer below the soil surface, impacting total
481 solute transfer into surface runoff. The random disposal of Cr (VI) wastes formed chromium-
482 containing slag, and the rainfall causes their movement (Ye et al., 2019). Rainfall intensity
483 significantly affected the migration and diffusion behaviour of Cr (VI). Heavy rainfall would bring
484 greater vertical transport risk, while weak rainfall conditions were more conducive to the reduction
485 of Cr (VI). The infiltration of rainwater through COPR sites, landfills and other waste sites
486 containing Cr, result in the infiltration of Cr leachate into the aquifers. If further disposal of Cr in
487 dump sites is stopped after initial dumping but due to the periodic rainfall the infiltration of Cr
488 contamination is continuous through the soil-groundwater system. The Cr moves from the upper
489 layer to the subsequent lower layer as a result, concentration of contaminants decrease in the top
490 layer but increase in the downward direction.

Zhang et al. (2022) studied effects of simulated acid rain and colloidal on fate and transport of Cr in vadose zone. Chromium transport in vadose zone soils (humic acid + cinnamon, montmorillonite + silt) during acid rain simulated in column experiments. Results indicate minimal Mt colloid impact on Cr; predominant Cr fixation in cinnamon soil with HA boosts adsorption by 12.8%. In the HA-Cr system, SO₄²⁻ inhibits adsorption, lowering efficiency from 31.4% to 24.4%. Guo et al. (2021) found that the combined effect of dispersion and rainfall caused the Cr concentration. Cr (III) and Cr (VI) temporal and spatial distributions are regulated by anthropogenic (location and emissions) and hydrologic factors such as water flow and particle transport. (Szalinska et al., 2010). Wu et al. (2022) studied the Cr (VI) transfer to the surface runoff from the soil. The soil adsorption properties and the θ₀ had the minimum effects on runoff Cr (VI). Results suggest that focusing on the analysis of precipitation conditions and soil parameters that determine the thickness of the mixing layer and the degree of mixing is critical for reducing pollution migration in surface runoff.

504 **5. Remedial Techniques for Chromium**

505 Soil-water systems can potentially contain elevated concentrations of chromium (Cr),
506 which can manifest as a significant threat due to its ability for inducing severe toxicity in humans,
507 plants, and animals (Escudero et al. 2017). Therefore, the remediation of soil contaminated with
508 Cr represents a critical research priority. Numerous remediation strategies have been developed
509 with the primary objective of mitigating chromium (Cr) contamination risks, diminishing land
510 fragmentation, facilitating land reclamation, and thereby fostering the expansion of agricultural
511 land to ensure long-term sustainability. Our present review focuses on the best demonstrated
512 available technologies which include bioremediation, phytoremediation, and physical remediation.

513 **5.1 Chemical Remediation of Chromium**

514 Numerous reducing agents, typically employed in acidic environments, have been utilized
515 for the remediation of Cr (VI)-contaminated soil-water systems, facilitating the conversion of Cr
516 (VI) to the Cr (III) state. In the remediation process of Cr (VI)-contaminated soil-water by
517 nanoscale zero-valent iron (nZVI), various modified materials such as CMC-stabilized nZVI,
518 nZVI@VR, and nZVI-RBC are commonly employed to prevent iron agglomeration and enhance
519 the efficiency of remediation. Cr (VI) adsorbs onto nZVI via surface pores and oxygen-containing
520 groups, leading to a direct reaction on the surface and reduced to Cr (III), while concurrently nZVI
521 undergoes oxidation to Fe²⁺ and indirect reduction of Cr (VI) occurs (Liu et al., 2020). Katsoyannis
522 et al. (2020) indicated the presence of an autocatalytic effect, where the concentration of Cr (VI)
523 influences its own reduction by ferrous iron (Fe (II)). Furthermore, the experiment suggested that
524 multiple additions of Fe (II) into Cr (VI)-spiked water are more effective in facilitating the
525 reduction process compared to adding all the required Fe (II) at once. In the study by Wazne et al.
526 (2007), the remediation of COPR involved the use of ferrous sulfate and calcium polysulfide. The
527 findings revealed that to meet the regulatory limits set by NJDEP (240 mg/kg for Cr (VI)) and

528 EPA TCLP (5 mg/L for Cr) after one month of curing. Fu et al. (2021), developed a remediation
529 method using chemical reduction with calcium polysulfide combined with microbial stabilization
530 to address elevated levels of Cr (VI), and reported 97% conversion rate of Cr (VI) to Cr (III).

531 **5.2 Bioremediation of Chromium**

532 Bioremediation of Cr remediation are found to be more prominent, which involve the use
533 of different biological mechanisms and processes. Bioremediation, the process by which living
534 organisms are either wholly or partially employed for the treatment of pollutants, represents a
535 pivotal approach in controlling environmental pollution caused by heavy metals like Cr (Elahi and
536 Rehman, 2019). Bioremediation technology offers several advantages over other treatment
537 processes, particularly when implemented on a large scale. These advantages include enhanced
538 efficiency, specificity, and environmental compatibility (Banerjee et al., 2019). Different species
539 of microorganisms may have distinct mechanisms for the removal and immobilization of Cr (VI).
540 In particular, Fungi can mitigate the harmful effects of biologically active, free potentially toxic
541 elements (PTE) through in vivo metal binding characterized by both high specificity and affinity
542 (Herath et al., 2014). Bacteria possess the capacity to alter the valence state of chromium from Cr
543 (VI) to mitigate its ecological toxicity (Elangovan et al., 2010).

544 Long et al. (2023) used *Penicillium oxalicum SL2* for bioremediation of Cr (VI)
545 contaminated soil. Authors conducted batch experiments with *Penicillium oxalicum SL2*, finding
546 that oxalic acid altered pH, favoring SL2 growth for Cr (VI) bioremediation in alkaline soil. The
547 results indicate oxalic acid enhances strain SL2's bioremediation efficiency by promoting
548 chromium bioleaching from soil and enhancing Cr (VI) removal in the leachate. Li et al. (2020)
549 utilized goethite (FeOOH) in conjunction with *Pseudomonas aeruginosa* (*P. aeruginosa*) for the Cr
550 (VI) bioremediation. Cr adsorption by FeOOH and FeOOH P. was monolayer. Extracellular
551 polymeric substances (EPS) produced by the bacterial strain and the leaching of Fe (II) from
552 goethite are essential contributors to the reduction of Cr (VI). Bansal et al. (2019) investigated the
553 dominance of *Pseudomonas aeruginosa*, *Serratia marescens*, *Alcaligenes faecal*, and *Klebsiella*
554 *oxytoca* in aerobic and anaerobic microbial cultures. They observed that they can reduce Cr (VI)
555 at concentrations of up to 50 mg/L in 6 hours under aerobic situations and 24 hours under anaerobic
556 situations. Both under aerobic and anaerobic conditions, the presence of Fe (III) had no effect on
557 the reduction of Cr (VI). Raman et al. (2017) suggested that *S. maltophilia* SRS₀₅ can be used for
558 bioremediation of Cr as a promising agent. Jeyasingh and Philip (2005) investigated Cr (VI) soil
559 bioremediation by an unidentified bacterial strain, achieving complete reduction of 1100 mg/kg
560 Cr (VI) in 20 days.

561 **Algae: Depending on the joining characteristics of the functional category and the**
562 **character of the operating conditions, several strategies for the minimization and elimination**
563 **of Cr (VI) through microalgae have been proposed. Biosorption of Cr (VI) onto extracellular**
564 **polymeric substances is the major mechanism responsible for chromium bioremediation by**

565 **Phaeodactylum tricornutum and Navicula pelliculosa (Hedayatkhan et al. 2018).** The majority
566 of Cr (VI) was collected in the cytoplasm, organelles, granules, heat-stable peptides, and proteins
567 of microalgal cells (Aharchaou et al., 2017). Furthermore, Leong et al. (2020) proposed that
568 microalgae strains possess bioaccumulation capabilities, engaging various mechanisms including
569 extracellular adsorption, reduction, volatilization, complex formation, ion exchange, intracellular
570 accumulation, chelation, and bio-methylation in the process of bioremediation.

571 Many microalgae species exhibit tolerance to and the ability to accumulate high
572 concentrations of Cr(VI). *Durvillaea antarctica* can remove 102.72 mg/g of Cr after 7 hours of
573 contact time. (Guarín-Romero et al. 2019). *Enteromorpha intestinalis* can remove up to 93.38 %
574 of Cr in 38.5 minutes by biosorption mechanism (Hamouda et al., 2020). *Pseudochlorella*
575 *pringsheimii* and *Chlorella vulgaris* can remediate 80% of Cr contamination in 1 day by
576 Bioaccumulation mechanism (Saranya and Shanthakumar, 2019). *Chlorella sorokiniana*
577 demonstrates the ability to withstand concentrations of up to 100 ppm Cr (VI) for three days,
578 achieving removal efficiency of up to 99.7% within 24 hours of contact time. *Pediastrum duplex*
579 and *pseudoanabeane mucicola* have respective tolerance limits for Cr (VI) of 1.936 and 0.224 g/L,
580 while the former has a removal efficiency for Cr of 71% (Dao and Vo, 2018).

581 **Fungi: Fungi are employed as highly adaptable biosorbents for Cr (VI) due to their**
582 **versatility, resilience in hostile conditions, and capacity to endure Cr (VI) concentrations**
583 **exceeding 10,000 mg/L (Vendruscolo et al, 2017). Fungal species typically exhibit resistance**
584 **to high metal ion concentrations (Bolisetty et al. 2019).** Fungi such as *Aspergillus oryzae*,
585 *Rhizomucor sp.*, *Aspergillus niger*, *Aspergillus carbonarius*, *Candida tropicalis*, *Trichoderma sp.*,
586 *Fusarium oxysporum* and *Penicillium chrysogenum* (Ashe and Duvnjak 1995; Igwe and Abia
587 2006; Srivastava and Thakur 2006; Vankar and Bajpai 2008; Elahi and Rehman 2017) have
588 demonstrated highly efficient biosorption capabilities for Cr(VI). For bioremediation of Cr (VI),
589 Bibi et al. (2108) analyzed endophytic fungi with four isolated strains namely *Aspergillus*
590 *fumigatus*, *Rhizopus sp.*, *Penicillium radicum* and *Fusarium proliferatum*. It effectively detoxified
591 up to 95% of Cr extracellularly. Kumar and Dwivedi (2019) demonstrated the effectiveness of the
592 fungus *Trichoderma lixii CR700* in bioremediating Cr (VI), achieving a remarkable 99.4%
593 reduction within 120 hours at a concentration of 50 mg/L of Cr (VI). Sanchez et al. (2019)
594 conducted batch experiments using fungal biomass of *Rhizopus sp.* to remove Cr (VI) ions from
595 aqueous solutions, finding that the maximum Cr (VI) uptake (99%) occurred at pH 2.0,
596 demonstrating its optimal conditions for Cr (VI) removal.

597 **5.3 Phytoremediation of Chromium**

598 Phytoremediation, a cost-effective and environmentally friendly technique, utilizes plants
599 and associated microorganisms to in-situ degrade or reduce the concentration of contaminants in
600 soil-water system, effectively addressing concentration of the toxic effects of contaminants
601 (Mohanty and Patra, 2012; Ali et. al., 2012; Yang et al., 2022; Ullah et al., 2023). The method is

602 energy-efficient, economical, ecofriendly, and it may be used with other remedial methods
603 (Suresh, Avishankar, 2004 and Ali and Sajid, 2012). Phytoremediation stands as an advancing
604 technology in the domain of soil-water remediation, with multiple advantageous attributes and
605 sustained applicability (Kassaye et al., 2017). Jia et al. (2022) developed an improved indicator
606 system for phytoremediator screening strategy and identified *Pennisetum* is an effective
607 phytoremediator for Cr. The author's belief is that *Pennisetum* holds significant promise for
608 extensive utilization in the remediation of chromium (Cr)-contaminated soils in the foreseeable
609 future. Sajad et al. (2020) analyzed 61 plants from 30 different families for concentration of Cr.
610 The Bioconcentration Factor (BCF), Translocation Factor (TF), and Biological Accumulation
611 Coefficient (BAC) were used to assess the plants' phytoremediation potential. Based on BCF value,
612 38 plant species show viability for the phyto-stabilization of Cr excluders. As indicated by
613 Saravanan et al. (2019), this method stands out as one of the best approaches compared to
614 conventional methods for remediating contaminated sites, as it leverages a diverse range of plant
615 species for effective treatment.

616 Phytoremediation encompasses distinct mechanisms contingent upon the nature of
617 contaminants (Foadi, 2020). Phyto-filtration (Hanus-Fajerska, 2016); phytoextraction (Bhargava,
618 2012); phytovolatilization (Jhilta et al., 2022); phytodegradation (Vishnoi and Srivastava, 2008);
619 phytodesalination (Vaudry et al., 2023); rhizodegradation (Sharma et al., 2022) and
620 phytodesalination (Zorrig et al., 2012). For phytoremediation nearly 500 plant species belonging
621 to more than 45 families have been identified till now (Srivastava et al., 2021). Most of the tolerant
622 hyper-accumulator plants changed hazardous metals into less hazardous and immobile forms
623 (Cervantes et al., 2001). Several plants such as *Lonicera japonica* Thunb (Meng et al., 2019);
624 macrophyte *Callitrichie cophocarpa* (Kyzioł-Komosi' et al., 2018), *Vigna unguiculata* (Eze et al.,
625 2018), *Spirodela polyrrhiza* (Singh and Malviya, 2019), *Cajanus cajan* (Jerez et al., 2016), *Canna*
626 *indica* L., *Typha angustifolia* L. (Taufikurahman et al., 2019).

627 **5.4 Permeable Reactive Barrier**

628 PRBs have rapidly developed in the past decade and are considered a highly promising in-
629 situ remediation technology. This is primarily because of their cost-effectiveness and their ability
630 to effectively immobilize various contaminants, making them suitable for a wide range of
631 applications. McMurtry and Elton (1985) gave the very first conceptual idea about PRB which
632 revolved around three main constituents including treatment methodology, the hydraulic data and
633 the geotechnical designs. After that it was first implemented in 1994 as a passive groundwater
634 remediation method for contamination of chlorinated ethylene in Base Borden, Ontario (Powell et
635 al., 1998). PRBs consist of filled materials like adsorbents and reducing agents, strategically placed
636 across contaminant plume flow paths. These barriers intercept and degrade various contaminants
637 while allowing groundwater to pass through. When considering the reactive media for PRBs, it is
638 important to consider their ability to immobilize contaminants as well as permeability. Adsorbents

639 are the preferred selection for PRBs due to their high adsorption capacity, chemical stability,
640 diverse pore structure, and the absence of secondary pollution.

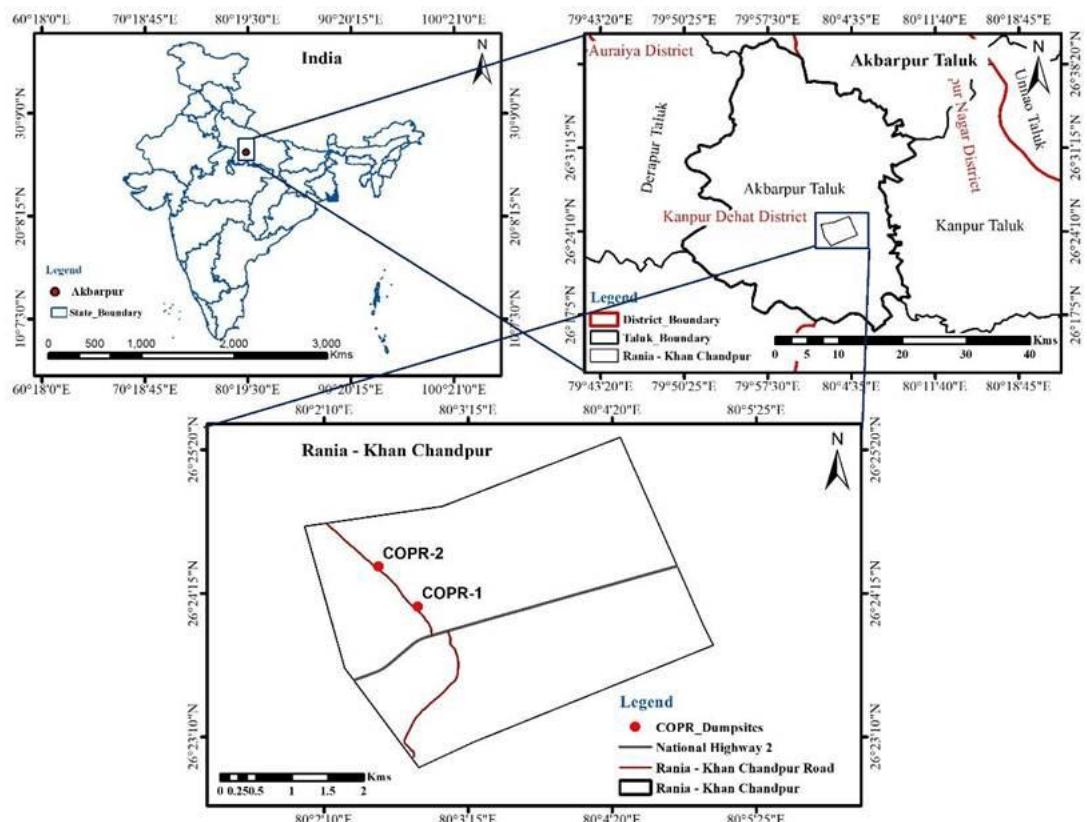
641 The primary advantage of PRBs over traditional Pump and Treat (P&T) systems is it
642 reduced demand for labour and energy during treatment. To design PRBs effectively, it is
643 important to understand the remediation mechanism. This mechanism involves the transport of Cr
644 contaminants through the barrier via advection, diffusion, and dispersion processes. Advection is
645 an important component of hydrological systems, while Darcy's law describes the horizontal
646 migration of pollutants in the direction of water flow through porous media. Fick's law governs
647 the diffusion of pollutants from areas of high concentration to low concentration, with
648 contaminant-porous media interactions playing a role in limiting diffusion (Vukojević Medvidović
649 et al., 2018).

650 A PRB operates based on two fundamental treatment principles: immobilization and
651 transformation. In the immobilization process, Cr contaminants either adhere to or form solid
652 precipitates within the reactive zone without any change in their chemical state. This adherence or
653 sorption occurs on the surface of the reactive materials, as adsorbents. Further, Cr contaminants
654 may precipitate by converting substances from a dissolved state into a solid state in the
655 contaminated solution (Scherer et al., 2020). The transformation process involves chemical
656 reactions that degrade the Cr contaminants into a non-harmful substance. The advantage of a
657 transformation process is that it eliminates the need for a comprehensive remediation of pollutants
658 (Thakur et al., 2020).

659 **6. Case Study of Cr Contaminated Rania and Khan Chandpur Villages**

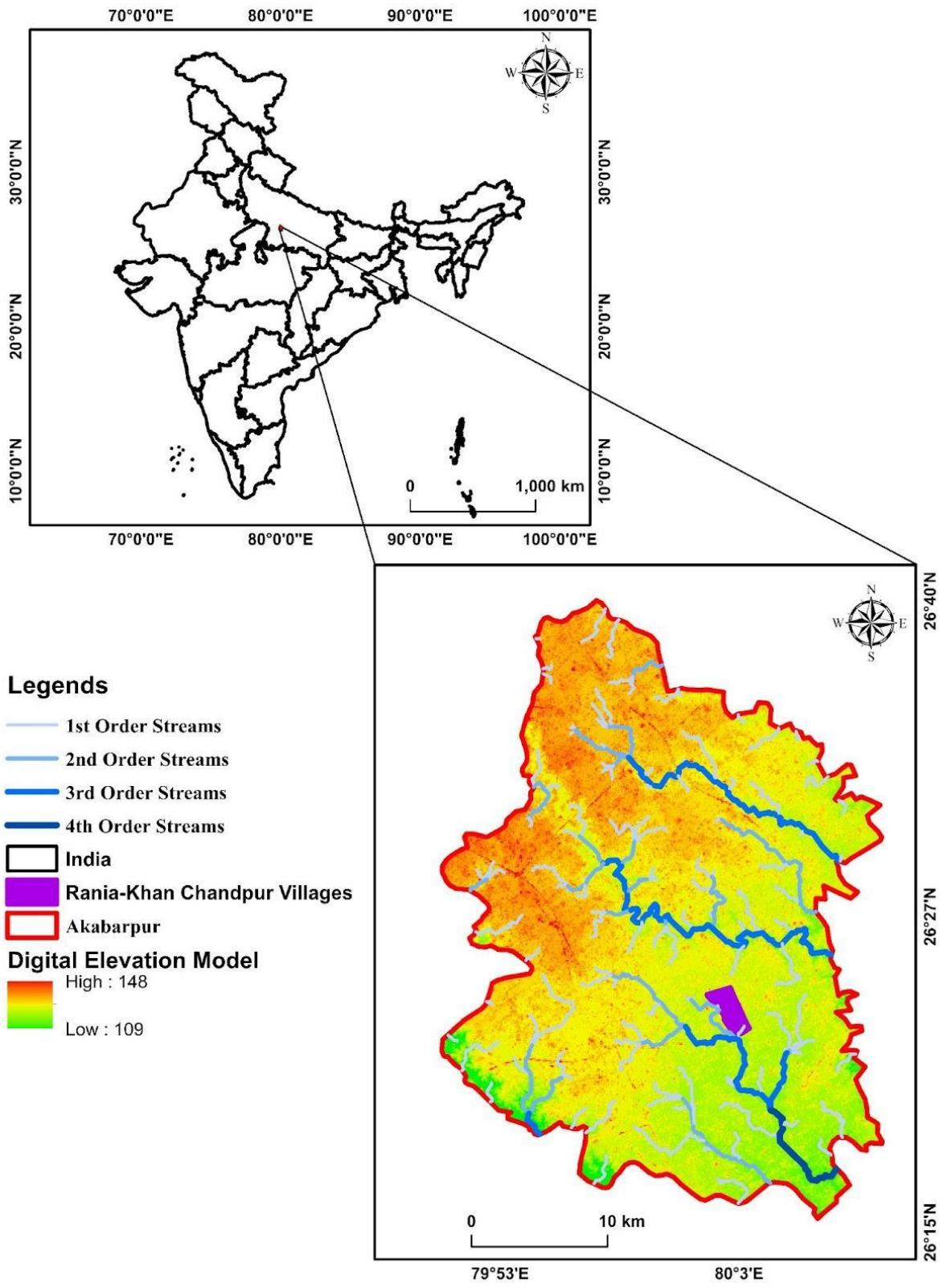
660 **6.1 Study Area**

661 The study was conducted in Rania-Khan Chandpur villages, situated within Akbarpur
662 Tahsil of Kanpur Dehat District, Uttar Pradesh State, India, approximately 30 Km away from
663 Kanpur city (Fig. 3). These villages are located near the NH-2 toll plaza, which is around 10 Km
664 from the main nearby city, Akbarpur. This region has humid subtropical climate conditions with
665 an average rainfall of 782.8 mm (Srivastava et al. 2013) from which about 70–80% of the total
666 precipitation occurs in July, August, and September during the monsoon season (Matern et al.
667 2017). In Rania, two COPR dump sites, totalling between 30,000 to 35,000 metric tons, were
668 identified by the Central Pollution Control Board. These sites served as repositories for COPR
669 between 1980 and 2005, remaining uncovered and exposed to the atmosphere until October 2022.
670 The COPR sites in Rania underwent capping, however, the COPR is continually leaching from
671 that site to the adjacent landscape. In this study COPR#1 is used for large COPR dumpsite and
672 COPR#2 is used for a compatible COPR dumpsite.



673

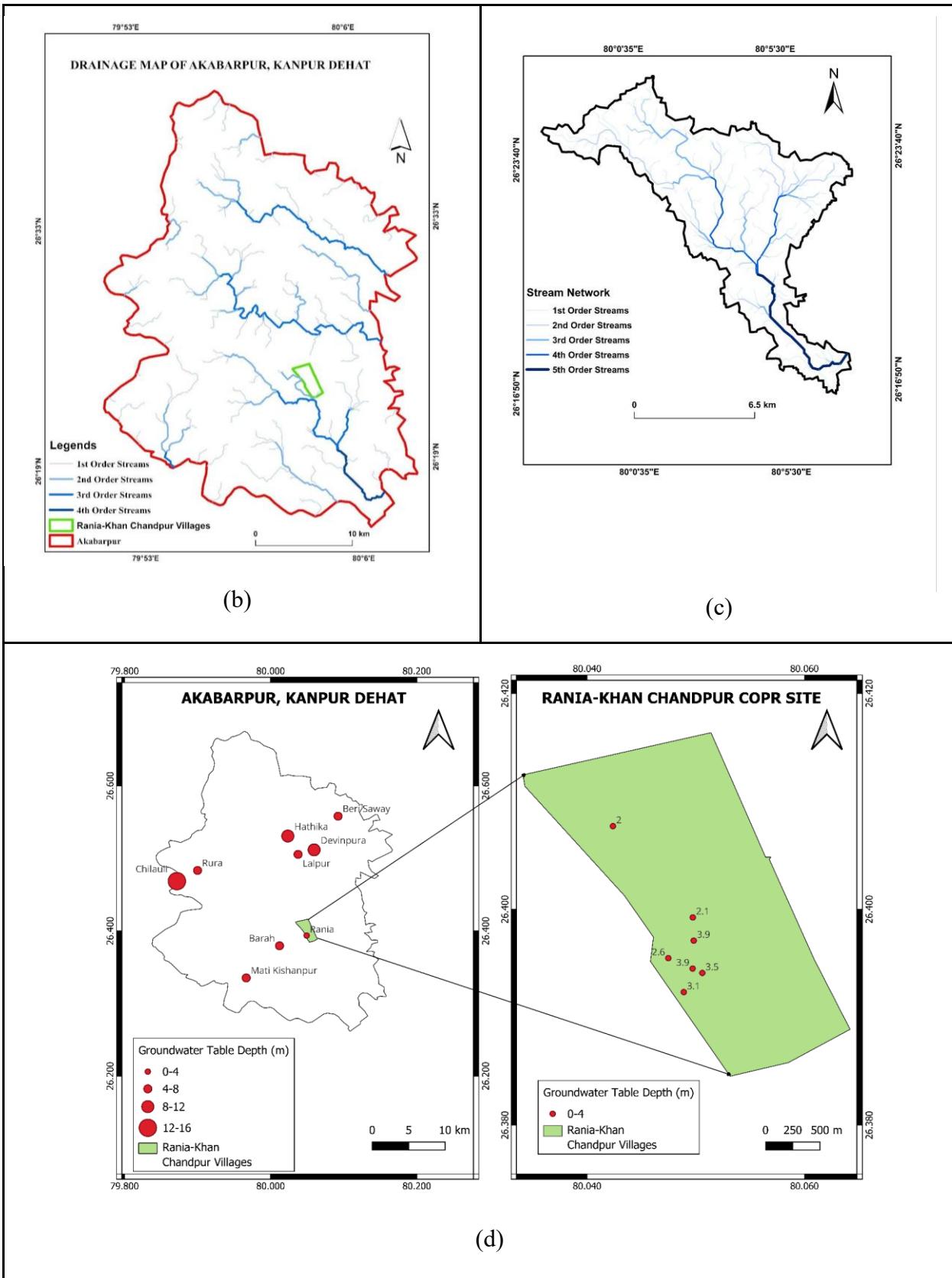
674 Figure 3: India map showing the location of Kanpur Dehat which extend the Akbarpur Tahsil with
 675 a focus on the Rania-Khan Chandpur Villages. Within this area the map indicates the location of
 676 two COPR dump sites COPR-1 and COPR-2, positioned near Rania adjacent to the Rania-Khan
 677 Chandpur Road.

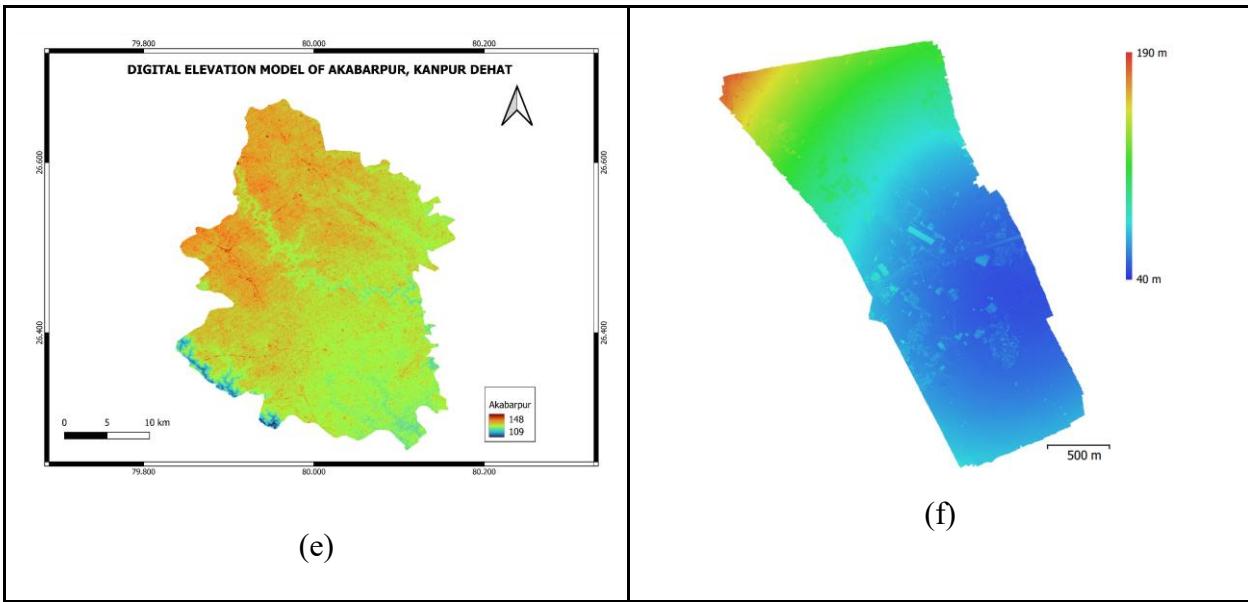


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(a)





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681 Figure 4. (a) Digital elevation model (b) Drainage map of Akbarpur, Kanpur Dehat (c) Drainage
 682 map of the study area (d) Groundwater table depth of Akbarpur, Khan Chandpur area (e) Digital
 683 elevation model of Akbarpur (f) Digital elevation model of Rania Khan Chandpur.

684 **6.2 Research Gaps**

685 Cr leachate from the COPR can infiltrate in the vadose zone and ultimately meet the
 686 (shallow-) groundwater which can result as huge Cr contamination in the subsurface. Our initial
 687 (Gupta et al. 2023a; Deoli et al. 2023) and other's works (Singh et. al., 2009; Matern et. al., 2017)
 688 have found that groundwater, surface-water, and soil samples from the past have shown signs of a
 689 Cr emanating from the COPR dump site in and around Rania-Khan Chandpur villages. In the
 690 subsurface, Cr can be in the form of Cr (III) or Cr (VI), the mechanisms and kinetics of Cr release
 691 are obscure (Bhattacharya et. al., 2019). The release of Cr from the COPR dump site can be
 692 influenced by factors such as the pH of the water/soil, the presence of other chemicals or ions, the
 693 hydrogeology and hydrology of the site, the above and below-ground ecology and microbial
 694 community dynamics, and so on. Further, climate change may influence the fate and mobility of
 695 the Cr by altering the hydro-metrical and subsurface conditions. A better understanding is
 696 needed to improve remediation practices including evaluation of potential Cr transport pathways
 697 and rates, and more efficient clean-up strategies. However, there remains a dearth of knowledge
 698 on Cr fate, transport, and remediation in the soil-water system (Table 6).

699 Considering the research gaps prior to the implementation of this research work, a series
 700 of investigations has been initiated to generate baseline data of soil-water quality at the Rania-
 701 Khan Chandpur site. This has resulted in basic knowledge of the initial site conditions and

702 profusion of excellent baseline data facilitating a better understanding of the soil-water system at
 703 this site. Photogrammetric approaches based on UAV surveys have seen significant developments
 704 throughout the past decade. Aerial imaging was performed at Rania-Khan Chandpur site in
 705 November 2022. The camera model ILCE-5100 has been used in UAV surveys with resolution of
 706 6000×4000 , 16 mm focal length and $4 \times 4 \mu\text{m}$ pixel size. A total of 2908 images have been taken
 707 from the flying altitude of 126 m. The ground resolution was $2.79 \text{ cm pixel}^{-1}$ and the total coverage
 708 area was 4.69 km^2 . The DEM and other maps have been shown in Figure 4. Drone images often
 709 have distortions due to the drone tilting from wind or movement, as well as issues from the camera
 710 and lens used. Therefore, these images need to be corrected to fix these distortions. In this study,
 711 the projection was 24363038 and the reprojection error was 1.37 pixel.

712 Table 6: Summary of Studies performed to investigate Cr contamination in Rania-Khan Chandpur
 713 Villages

Citation	Lat/long	Scale of Study	Samples/Environmental Conditions	Methods	Key Observations
Singh et al. (2012)	Rania ($26^{\circ}24'12.4''$ N, $80^{\circ}02'51.2''$ E)	Lab Scale	27 soil samples were collected from different depths varying from 0 to 40 feet.	nZVI used in this study was synthesized in the laboratory by the reduction of FeCl_3 with NaBH_4 . Metal analysis and batch experiments.	five samples had shown significant level of Cr (VI) with an average concentration of 15.84 mg Kg^{-1} .
Foldi et al. (2013)	Rania ($26^{\circ}24'12.4''$ N, $80^{\circ}02'51.2''$ E)	Lab Scale	Total 21 soil samples were collected from 0 to cm below the surface.	<ul style="list-style-type: none"> C, N and S were determined by elemental analyzer. Other elements were analyzed by XRF. X-ray diffraction is used for mineralogical composition. Batch experiment has been performed to know the Cr (VI) solubility and deduce processes 	<ul style="list-style-type: none"> The COPR displays high Fe and low Mg contain which can be referred as ferro chromite or (high-)ferric (magnesio-)chromite. Total Cr contents of highly contaminated material ranging from 65.7 to 110 g/kg. Dissolution was responsible for regulating the Cr (VI) concentrations in the eluates. With over 1400 mm (90%) of rainfall occurring during the monsoon season, there is a

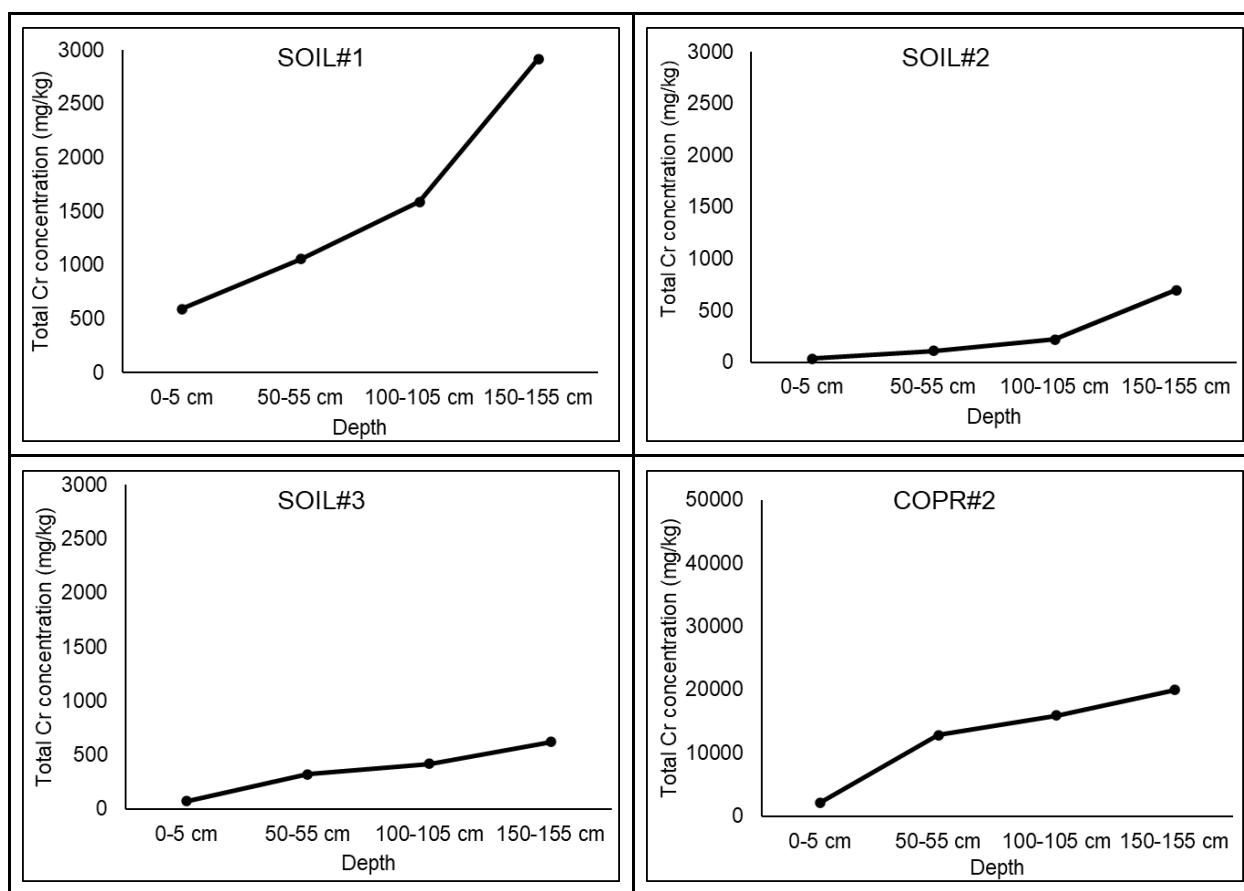
				of solubility-controlling.	higher chance of increased mobilization of Cr (VI) due to a wider ratio of COPR to percolating rainwater.
Prakash et al. (2011)	Rania ($26^{\circ}22'00''$ N to $26^{\circ}25'00''$ N and $79^{\circ}58'00''$ E to $80^{\circ}07'00''$ E)	Lab Scale	<ul style="list-style-type: none"> A total of 20 water samples were collected. The elevation of the land surface varies from 117 m to 139 m. The average annual rainfall of the district was 782.8 mm. 	<ul style="list-style-type: none"> All the heavy metals were tested by standard water quality method. 	<ul style="list-style-type: none"> Cr in collected groundwater samples found up to 42.45 mg/l. Iron concentration in collected samples was also more than the permissible limit up to 2.629 mg/l. Periodic monitoring of water quality mainly for Cr is very essential for Rania, Kanpur.
Matern et. al. (2016)	Rania ($26^{\circ}24'12.4''$ N, $80^{\circ}02'51.2''$ E)	Field and Lab Scale	<ul style="list-style-type: none"> 57 groundwater samples were in pre monsoon and 70 after monsoon. Rainfall: 681 mm average rainfall from 2009 to 2013. Aquifer system: Phreatic zone depth: 25 to 130 m. Middle aquifer: 90 to 240 m. Lower aquifer: 180 to 450 m. 	<ul style="list-style-type: none"> Standard Water Quality Analysis Methods. IBM SPSS Statistics was used for statistical data analysis. 	<ul style="list-style-type: none"> Cr (VI) concentration in groundwater were <0.005 to 34.8 mg/l for Rania, <0.005 mg/l to 115 mg/l for Chhiwali and <0.005 to 2.0 mg/l for Godhrauli. 95% of the groundwater samples are unsafe for drinking. COPR dumps are the main source of Cr (VI) in groundwater. Urgent need of remediation for COPR
Matern et. al. (2016)	Rania ($26^{\circ}24'12.4''$ N, $80^{\circ}02'51.2''$ E) and Chhiwali ($26^{\circ}11'45.8''$ N, $80^{\circ}32'21.3''$)	Lab Scale	COPR Samples	<ul style="list-style-type: none"> Lasere particle analyzer was used for particle size distribution. Total C, N and S were determined by CNS analyzer. Mineralogical composition was 	<ul style="list-style-type: none"> both samples had a pH of 12.4, which is due to lime (CaO) during the roasting process. The two COPR samples had total Cr levels of 81 and 74 g/kg. where 80 to 87% of the Cr was found as Cr (III).

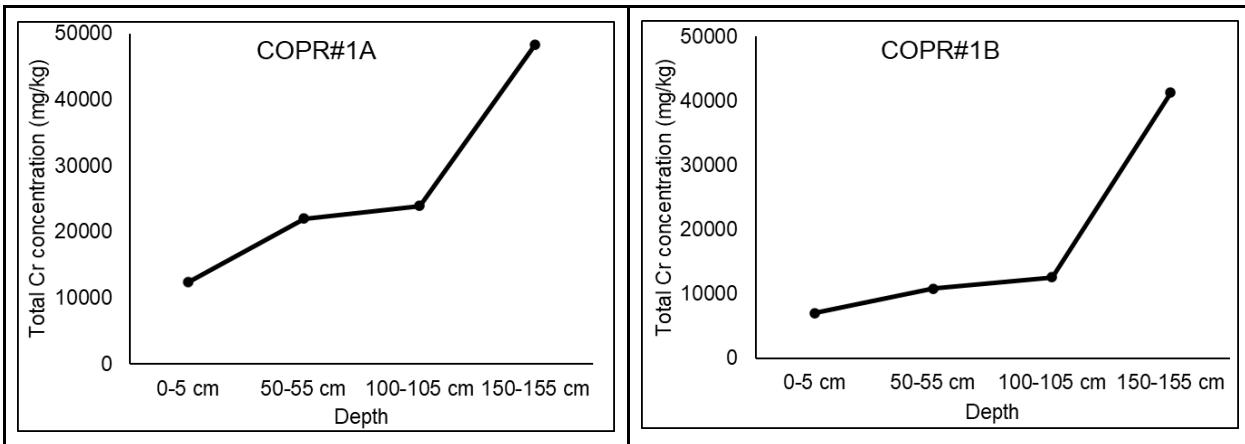
	E), Kanpur India			analyzed by DEM analyses.	<ul style="list-style-type: none"> Two new minerals in Indian COPR i.e., voltaite and grimaldiite.
Matern et al. (2020)	Rania (26°24'12.4" N, 80°02'51.2" E) and Chhiwali (26°11'45.8" N, 80°32'21.3" E), Kanpur India	Lab Scale	COPR collected samples after monsoon	<ul style="list-style-type: none"> Column experiment performed by a column of 20 cm long and 4 cm inner diameter. Cr (VI) concentration was determined by spectrophotometer at 540 nm. Geochemical modelling done by Visual Minteq 3.0. 	<ul style="list-style-type: none"> Cr (VI) in both COPR sites is highly soluble and is a major component of leachate. Cr (VI) will continue to be released from the COPR dumpsites. During the column experiment, the ratio of Cr (VI) to total Cr decreased over time.
Kumar et al. (2023)	Rania (26°24'57" N, 80° 3' 28" E)	Lab ad Field Scale	<ul style="list-style-type: none"> 54 BCS sludge samples were collected from 15 different locations during ore and post monsoon season. The samples were collected from 3 different depth: Top surface (0-15 cm), subsurface layer (15-75 cm) and bottom layer (75-150 cm). 	<ul style="list-style-type: none"> Toxicity characteristic leaching procedure (TCLP) based concentration of Cr were assessed. Cr-VI concentrations using spectrophotometrically at 540 nm. pH, EC, and TDS using standard methods. Organic matter by Walkley-Black procedures. 	<ul style="list-style-type: none"> Concentration of total and TCLP based Cr and Cr (VI) increase with increase in sample depth. Highly leaching tendency of sludge during both seasons. Illegally dumped require proper treatment, storage, and disposal facilities. The leaching characteristic of salts tends to increase as the depth level of the dumpsite increases.

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715 6.3 Cr in Soil Samples

716 For sampling of soil and COPR from hazardous waste, the standard method was followed by
717 CPCB 2011. The samples were collected in zipper polyethene bags which were air dried in the
718 laboratory. All samples were air-dried, and the concentrations of total chromium. Collected
719 samples were digested in Aqua regia solution (3:1: hydrochloric acid: nitric acid) and filtered using
720 0.2 μ syringe filters (Axiva, Cat No. SFNY25RB) and stored in the cold containers in the
721 laboratory. All samples were analysed for Cr using Inductively Coupled Plasma Mass
722 Spectrometry (ICP-MS) with equipment from Agilent Technologies, Singapore, specifically the
723 ICP-MS 7900 model within 72 hours.





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Figure 5: Cr contamination in Soil and COPR samples at different depths

726 Total Cr concentration in soil and COPR samples increases with an increase in depth. The top
 727 layer (0–5 cm bgl) was 12373, 7000, and 2134 mg kg⁻¹ in cores collected from the northern
 728 boundary of main dump site (COPR#1), middle of the main dump site (COPR#2) and small dump
 729 site (COPR#3) respectively, which was increased to 21996, 10786 and 12789 mg kg⁻¹ in samples
 730 collected from 50-55cm bgl in the northern boundary of the main dump site, middle of the main
 731 dump site and small dump site, respectively. The total Cr concentrations were further increased at
 732 100–105 cm depth as 23958, 12591, and 15930 mg kg⁻¹ in the northern boundary of the main
 733 dump site, middle of the main dump site and small dump site respectively. The total Cr
 734 concentrations were highest in the bottom layer of cores, 150-155 cm bgl, as 48330, 41274, and
 735 19930 mg kg⁻¹ in main Cr sites, middle of main COPR sites, and small dump sites samples,
 736 respectively, which was quite close to the values reported by Kumar et al. (2023) for 75-150cm
 737 bgl. Total Cr concentrations were increased with depth in COPR cores collected from three
 738 locations. Similar trends of total Cr concentrations were observed in the soil cores as well in which
 739 concentrations of total Cr were also increased with depth, but the ranges of total Cr concentrations
 740 (33.3-2918.8 mg kg⁻¹) in soils were much lower than whatever was observed in the COPR (2134-
 741 48330 mg kg⁻¹). The total Cr concentrations in soil cores were 590, 33, and 72 mg kg⁻¹ in top layer
 742 samples (0–5 cm bgl) from SOIL#1, SOIL#2, and SOIL#3, respectively. The highest total Cr
 743 concentrations were 2918, 695, and 618 mg kg⁻¹ in bottom layer samples (150–155 cm bgl) from
 744 SOIL#1, SOIL#2, and SOIL#3, respectively.

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746 **6.4 Cr in Groundwater Samples**

747 Total 78 groundwater samples were collected in October 2022 to know the status of Cr in and
748 around COPR sites in Rania and Khan Chandpur villages. The samples were collected from hand
749 pumps and each sample was collected after approximately 20 L of water was flushed out from this.
750 The samples were immediately kept in the cool box during transportation to the laboratory. All
751 samples were filtered by 0.45 μ syringe filters (Axiva, Cat No. SFNY13X). The analysis has been
752 done using ICP-MS (Agilent Technologies, Singapore, Model: ICP-MS 7900).

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