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Chromium contamination in paddy soil-rice systems and associated human health risks in Pakistan

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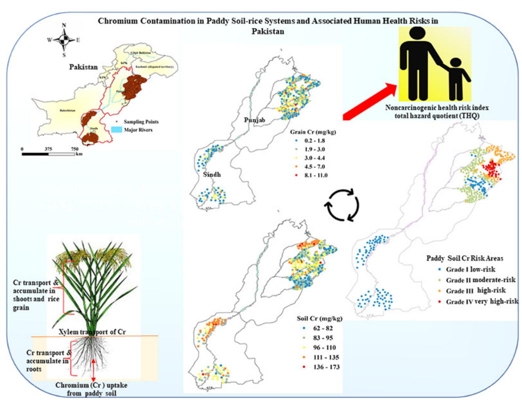
HIGHLIGHTS

- Chromium 97.4% rice grain samples exceeded the threshold 1.0 mg kg^{-1} from Pakistan.
- Chromium 62.6% soil samples exceeding the China natural background threshold 90 mg kg^{-1} and lower than the (pH-dependant > 7.5 thresholds 350 mg kg^{-1}.
- GWR model indicated the influencing independent variables on Cr accumulation in rice grain varies.
- Paddy soil samples exceeding Cr critical thresholds and loading capacity distributions confirm the risk of Cr in paddy soil.
- Total hazard quotient (THQ << 1) indicates no potential health risk originating from Cr exposure.

GRAPHICAL ABSTRACT



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ABSTRACT

Chromium (Cr) contamination in paddy soil-rice systems threatens human health through the food chain. This study used a new dataset of 500 paddy soil and plant tissue samples collected in the rice-growing regions of Sindh and Punjab Provinces of Pakistan. Overall, 97.4% of grain samples exceeded the Cr threshold values of 1.0 mg kg^{-1}, determined by the China National Food Standard (CNFS). The Cr in paddy soil, 62.6% samples exceeding the China natural background threshold value (90 mg kg^{-1}) for Cr concentration in paddy soil, and lower than the (pH-dependant > 7.5 threshold value for Cr 350 mg kg^{-1}) as determined by China Environmental Quality Standards (EQSs) for paddy soil (GB15618-2018). Geographically weighted regression (GWR) modelling showed spatially nonstationary correlations, confirming the heterogeneous relationship between dependent (rice grain Cr) and independent paddy soil (pH, SOM, and paddy soil Cr) and plant tissue variables (shoot Cr and root Cr) throughout the study area. The GWR model was then used to determine the critical threshold (CT) for the measured Cr concentrations in the paddy soil system. Overall, 38.4% of paddy soil samples exceeding CT values confirm that the paddy soil Cr risk prevails in the study area. Furthermore, the GWR model was applied to assess the loading capacity (LC), the difference between the CT, and the actual concentration of Cr in paddy soil. Loading capacity identified potential paddy soil Cr pollution risk to rice grain and assessed the risk areas. Overall LC% of samples paddy soil Cr risk areas grade: low-risk grade I (34.6%);

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moderate-risk grade II (15.8%); high-risk grade III (11.2%); and very high-risk grade IV (38.4%) have been assessed in the study area.
The human health index, total hazard quotient (THQ ≪ 1), indicates no potential health risk originating from Cr exposure to the population. However, the excess Cr level in paddy soil and rice grain is still a concern. The current study's results are also valuable for the national decision-making process regarding Cr contamination in the paddy soil-rice system.

1. Introduction

Heavy metal contamination in soil-plant systems has become a severe global concern due to its persistence, bioaccumulation, and toxic effects (Ali et al., 2020). Worldwide in the past five decades, the amount of Chromium (Cr) released into the environment has been assessed at more than 30,000 tons originating from diverse natural and anthropogenic activities, most of which significantly accumulate in industrial and agricultural soil, causing Cr pollution, have become increasingly important (Li et al., 2020; Shahid et al., 2017). Naturally, Cr mainly occurs in different environmental matrices, including soil, water, rocks, gases, and volcanic dust (Ertani et al., 2017; Shanker et al., 2005). Chromium is typically tightly bound to primary rock-derived phases (Quantin et al., 2008). Additionally, Cr can co-occur with amorphous Fe oxides (McClain et al., 2019). Chromium can exist in original minerals coprecipitated with iron (Fe), manganese (Mn), and aluminum (Al) oxides and hydroxides, which are usually adsorbed on soil particles and complexed with soil organic compounds (Hsu et al., 2015; Shahid et al., 2017). In contrast, Cr's primary anthropogenic sources in agricultural soil include sewage sludge, poultry or livestock manure, phosphate or compound fertilizers, and atmospheric deposition (Ma and Hooda, 2010; Shahid et al., 2017).

Chromium in polluted agricultural soils accumulates in the crop system; therefore, it carries human risks due to food chain contamination (Ahemad, 2015). Previous studies have reported that Cr contamination of paddy soil-rice systems is extensive in rice-growing countries, including Japan, India, Bangladesh, Iran, China, and Australia (Ali et al., 2020; Arunakumara et al., 2013). Elevated Cr concentrations in the paddy soil system cause increased Cr uptake, translocation, and accumulation, ultimately increasing Cr concentrations in paddy tissues (e.g., roots, shoots, leaves, and rice grains), which can threaten food security by reducing rice yields and quality due to the phytotoxic effects of Cr (Hu et al., 2019; Mahfooz et al., 2020; Shahid et al., 2017). Hence, numerous regulatory bodies in diverse countries such as the CNFS have strictly regulated the permissible concentration of Cr in paddy soil-rice systems. In rice grains, the maximum acceptable limits for Cr are 1.0 mg kg^{-1} (Wu, 2012). However, several recent publications on Environmental Quality Standards for Chinese paddy soil cite revised 2018 thresholds (GB15618-2018: Risk control standard for soil contamination of agricultural land) that range between 250 and 350 mg kg^{-1} (pH-dependent). Intervention values for Cr concentration in agricultural soil range from 800 to 1300 mg kg^{-1}, and the natural background threshold value set for Cr concentration in paddy soil is 90 mg kg^{-1} (Li et al., 2020; Sun et al., 2019).

In paddy soil system, the Cr availability is mainly associated with soil properties, such as pH, soil organic matter (SOM) content, soil cation exchange capacity (CEC), the redox potential (E_h), clay minerals contents, and Fe/Mn oxides (Ma and Hooda, 2010; Young, 2013). Among these, all soil properties, it was found that the pH has a significant role in determining Cr speciation bioavailability due to strong influence on solubility in the soil system, mainly in soil solution (Violante et al., 2010). In paddy soil systems, trivalent chromium Cr(III) and hexavalent chromium Cr(VI) are common and the most stable (Fan et al., 2020). Trivalent chromium and Cr(VI) differ in their transport and chemical behaviour in paddy soil systems (Hseu and Iizuka, 2013; Ma and Hooda, 2010). Paddy soils are typically flooded, conditions that favor the formation of reduced Fe [Fe(II)]. In the presence of Fe (II) or other reducing agents, like sulfides, Cr(VI) is rapidly reduced to Cr (III) and immobilized in soil (Fendorf et al., 2000). Thus, pH is the main factor that controls Cr speciation, bioavailability, and mobility in paddy soil systems (Xiao et al., 2021). In paddy soil, at a pH interval between 4 and 8, Cr(III) is most stable available in hydrolysed forms as $CrOH_2^+$, $Cr(OH)_2^+$, and $Cr(OH)_3$ (Ertani et al., 2017; Ma and Hooda, 2010). At a pH interval between 2 and 14, Cr(VI) is thermodynamically the most stable and available in forms as $HCrO_4^-$ and CrO_4^{2-} (Atiaga et al., 2021; Colombo et al., 2014; Ertani et al., 2017; Ma and Hooda, 2010). Apart from soil pH, the SOM also plays the main role in influencing the availability of Cr in the soil system; SOM can retain Cr in exchangeable form (Terzano et al., 2021). Furthermore, SOM in paddy soil solution can also act as chelates and increase Cr availability to the plant system (Zeng et al., 2011).

Paddy plants have high efficiency for the uptake, transport, and accumulation of Cr and its species Cr(III) and Cr(VI) (Rani et al., 2020) through particular transporters that absorb ions essential for the plant's metabolic activity (Singh et al., 2013). Trivalent chromium is mainly taken up and translocated in plants via a passive mechanism (Ali et al., 2020). In contrast, Cr(VI) uptake and translocation occur via an active mechanism, typically through sulfate or phosphate transporters, because of the structural resemblance of Cr(VI) to sulfate and phosphate (Shahid et al., 2017). The paddy soil properties, available Cr concentration, and rice varieties affect the uptake and accumulation of Cr in rice grains (Khan et al., 2013). Hence, for a particular rice type, the extent of Cr contamination risk in paddy soil systems can be determined by soil properties, such as the SOM and pH. In previous studies, the influences of paddy soil were infrequently measured in mapping the local-scale Cr contamination hazard in paddy soil systems in different countries. The combined impact of soil properties, i.e., pH and SOM that affect Cr phytoavailability in the paddy soil-rice systems, can be represented as nonstationary three-dimensional variables within a study area. This approach could support the development of spatially nonstationary paddy soil Cr limits that, in turn, ensure that the Cr in crops is below an acceptable level. Chromium in polluted soils accumulates in the crop system; therefore, it carries human risks due to food chain contamination (Ahemad, 2015).

Chromium can positively and negatively affect human health based on daily intake, exposure duration, and speciation. Historically, Cr(III) was considered essential, but recent literature has disproven its necessity for human nutrition (EFSA Panel on Dietetic Products and Allergies, 2014). Previously, for humans, Cr(III) suggested ideal daily intake to support protein, glucose, and fatty acid metabolism ranging between 50 and 200 μg day^{-1} (Ertani et al., 2017; WHO, 2000). However, the mechanism for these roles for Cr(III), an essential function in metabolism, has not been substantiated (EFSA Panel on Dietetic Products and Allergies, 2014). Hexavalent chromium is considered 10 to 100 times more toxic than Cr (III) due to its tendency to act as a strong oxidant (Ertani et al., 2017; Kim et al., 2002). Hexavalent chromium can cause severe human health effects, including skin eruptions, ulceration, bronchitis, asthma, gastric disturbances, genetic mutations, hepatic damage, kidney failure, and lung cancer (Ertani et al., 2017; Kotaś and Stasicka, 2000).

Rice is the most dominant agricultural product in Pakistan. This extensive research performed a national-scale assessment in rice-growing regions of Sindh and Punjab Provinces of Pakistan. Our key objectives were to 1) elucidate the Cr contamination status in the paddy soil-rice systems in Pakistan; 2) to understand the spatial variability and nonstationary regression relations among measured paddy soil (pH, SOM, paddy soil Cr) and plant tissue variables (shoot Cr, root Cr) with GWR modelling; 3) to generate spatial distribution maps of the paddy soil Cr critical threshold (CT) and loading capacity (LC) for crops in rice-growing areas; 4) to elucidate the risk areas where the paddy rice Cr concentration exceeds or might exceed the allowable threshold; and 5) to assess the health risks of Cr exposure through rice consumption based on our new data points. Furthermore, this study will aid in the development of specific measures to decrease Cr accumulation in rice systems by adjusting various paddy soil properties in the risk regions of the research area.

2. Materials and methods

2.1. Study area

Pakistan, located in South Asia, is surrounded by the Arabian Sea coastal belt. The country includes 137 administrative divisions/districts and is approximately 796,095 km^2 in area. One-third of the country is covered mainly by deserts, and the rest are generally agricultural lands and grasslands (Bhowmik et al., 2015). Pakistan's agricultural lands are typically within the flat, low-lying Indus Plain situated in eastern Pakistan. The Himalayan and Karakoram Mountain ranges and the Hindu Kush foothills are in the northern zone; the high mountain regions and Kashmir (disputed territory) are situated in the vast northwest Balochistan Plateau, which is located in the western part of the country (Fig. S1).

Pakistan's climatic conditions are generally semiarid to arid, except in the northwest temperate zone (Podgorski et al., 2017). Sand deposits and unconsolidated gravel at approximately 300 m depth occur in Quaternary alluvial deposits with truncated organic matter in soils located in the eastern part of the Indus Plain. The riverine systems include the Indus, Jhelum, Chenab, Sutlej, and Ravi Rivers in the northwest temperate zone (Ali et al., 2019b). Wind-blown sands are dominant in the adjoining regions, while porous gravels within a limited range are found in northwestern areas. Because of the accessibility of water resources and productive soils, the Indus Plain area, primarily in the Sindh and Punjab Provinces of Pakistan, includes extensive paddy and other agricultural production areas (Eqani et al., 2016; Podgorski et al., 2017).

2.2. Sample collection and preparation

In the present study, from September to October 2017, paddy soil and plant samples were taken from the rice-growing districts of the Sindh and Punjab Provinces during the rice harvest. In the study area of Sindh Province, the sampling districts included Thatta, Badin, Tando Allahyar, Hyderabad, Tando Mohammad Khan (TM Khan), Dadu, Larkana, and Shikarpur. In the study area of Punjab Province, the sampling districts included Bahawalnagar, Mindi Bahualdin, Sahiwal, Hafizabad, Gujrat, Gujranwala, Narowal Sialkot, Sheikhupura, Lahore, Faisalabad, Okara, Nankana Sahib, Kasur, Pakpattan, and Chinoat (Fig. S2). Both Provinces' study areas are within the Indus Plain region and cover approximately 6000 km^2. Several anthropogenic sources pollute these study areas, including brick kilns, wastewater effluent, chloralkaline plants, industrial areas, coal-fired power plants, incinerators, sewage sludge, poultry or livestock manure, phosphate or compound fertilizers, and atmospheric deposition (Ali et al., 2019a; Aslam et al., 2020). A total of 500 paddy soil (rhizosphere) and plant tissue (shoot, grain, and root) samples were randomly collected from 6 to 8 km^2 plots in pre-existing paddy fields in Punjab ($n = 414$) and Sindh ($n = 86$) (Fig. 1).

Each sample for the spatial distribution study was a composite of not less than five subsamples taken within 10 to 15 m of each specific sampling site. Paddy tissues and subsequent paddy soil samples (0 to 10 cm depth) were randomly collected from each area. Each paddy plant subsample was initially uprooted and carefully washed with paddy field irrigation water. The subsamples were mixed into composite samples, individually sealed in polyethylene bags, and transferred to clean/dry conditions. Simultaneously, paddy plant tissue (grain, shoot, and roots) samples were rewashed with ultrapure 18.2 MΩ tap water (Milli-Q® Integral system) and air-dried for several days at room temperature.

The sampled rice grains were parted into bran, husk, and polished rice. Next, the paddy plant tissue samples (polished rice, shoots, and roots) were powdered using a grinding machine (IKA®A11 basic analytical mill). Beforehand preparing every sample, the grinding instrument was carefully washed twice with 90% grade ethanol. All powdered samples were enclosed in polyethylene bags and stored at room temperature. The paddy soil samples were air-dried at room temperature, and each sample was divided into two parts. The 1st part of the sample was crushed using an agate motor grinder and passed through a nylon sieve (2.0 mm) mesh to measure soil pH. The 2nd part was ground and passed through a nylon sieve (100 mm) mesh to measure Cr wrapped in polythene bags. Soon after, all samples were moved to the State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences (SKLEG-IGCAS), Guiyang, China, for chemical analysis.

2.3. Chemical analysis

To measure soil pH, 4 g of soil was shaken in 10 mL of ultrapure water in a polytetrafluoroethylene (PTFE, Teflon) bomb (Rayment and Lyons, 2011) and analyzed using a PHEP-EC-N sensor (Hanna, Mauritius) with an accuracy of 0.001 pH units. At the same time, SOM was measured through the potassium bichromate wet combustion method (Wu et al., 2010). For the Cr in paddy soil, 50 mg (100 mesh) samples were digested with 3 mL of ultrapure nitric acid (HNO_3) and 0.5 mL of hydrofluoric acid (HF) in the oven for 48 h at 150 °C (Hu and Qi, 2014). A total of 100 mg of a powdered sample of paddy tissues (root, shoot, and grain) was digested with HNO_3 for 7 h at 150 °C in high-pressure digestion Teflon bomb. After digestion, the bombs were unscrewed and supplemented with 1 mL of 30% (v/v) ultrapure hydrogen peroxide (H_2O_2) and heated on a hot plate (110 °C) until the solution was almost completely evaporated (Chang et al., 2019). Then, 2 mL of HNO_3 and 3 mL of ultrapure water were added to each bomb and again placed in an oven for 7 h at 150 °C. Then, the containers were unscrewed, and 5 mL of ultrapure water was added to each bomb. Each sample was filtered through a 0.45 μm membrane (Whatman, USA). Finally, all diluted samples were moved to an inductively coupled plasma mass spectrometry (ICP-MS, Agilent 77,009, California, USA) laboratory for Cr analysis.

For quality control and quality assurance, the sample replicates, method blanks, standard reference materials (SRMs1640a, USA), and certified reference materials (CRMs) for paddy soil and tissue (GSS-5, GSB-11, and GBWE 100359) were analyzed after each 10th sample during sample preparation and chemical analysis. All uncertainties and constraints were determined and controlled using consistent laboratory replicate verifying the instruments' accuracy and calibration over consistent runs with SRM 1640a. The recoveries of relevant analytes ranged between 98.2 and 105.6%, and the recovery of SRM 1640a ranged between 89.6 and 112.8%. Duplicate samples' relative standard deviation (RSD) was within 10%.

2.4. Bioaccumulation and translocation factors

Bioaccumulation factors (BAFs) are an index of the capacity of plant tissues to accumulate a metal based on the metal concentration in the soil system (Li et al., 2014). The BAFs of Cr were measured for each rice sample to calculate bioaccumulation effects in rice from the uptake of Cr in the paddy soil system by using the following equation (Li et al., 2014):

$$BAF_{S_{tissue}} = C_{tissue}/C_{paddy\ soil} \quad (1)$$

where C_{tissue} is the Cr concentration of paddy tissues (root, shoot, and grain) and $C_{paddy\ soil}$ is the Cr concentration of equivalent rhizosphere soil. The BAFs $^{>}$ 1 indicated a high accumulation of metals in the tissues of the paddy plant system.

The translocation factor (TF) assessed the possible accumulation of metals in rice (Lu et al., 2018). This TF ratio specifies the paddy plant's ability to translocate metals from its roots to its aerial tissues (shoots and

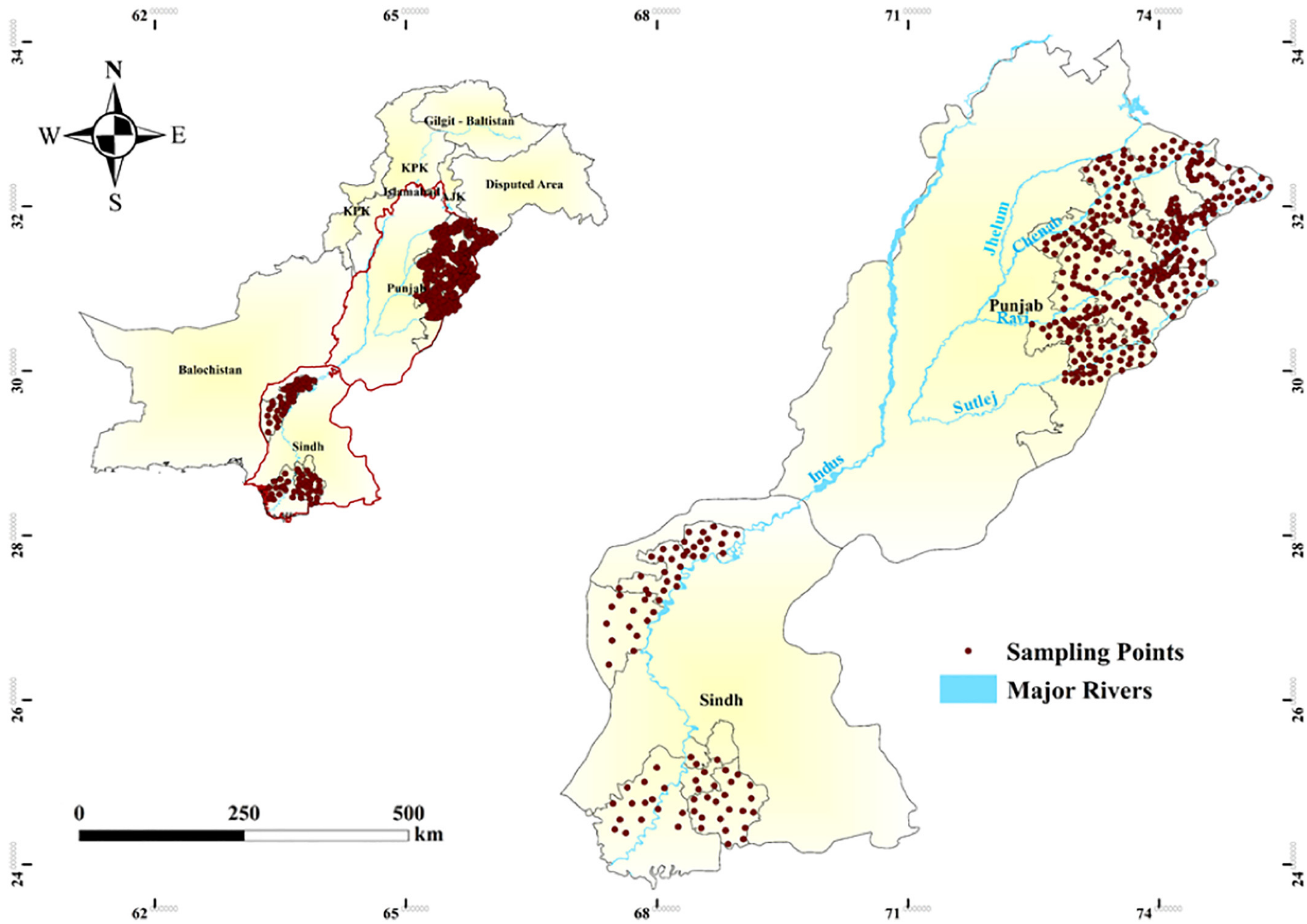

Fig. 1. Study area in Sindh and Punjab Provinces of Pakistan, with spatial distribution sampling of 500 data points.

grains). The TF ratio of Cr was calculated by the following equation (Li et al., 2014):

$$TF = C_{tissue\ 1}/C_{tissue\ 2} \tag{2}$$

where $C_{tissue\ 1}$ is the Cr concentration of paddy plant aerial tissue (shoot and grain) and $C_{tissue\ 2}$ is the Cr concentration of the corresponding underground paddy plant rice tissue (root). The TF values $^{<}$ 1 showed that the paddy accumulated the metal mainly stored in their roots, and TF $^{>}$ 1 suggested the translocation of trace elements to the aerial parts of the paddy (Li et al., 2014).

2.5. Geographically weighted regression modelling

Geographically weighted regression modelling is a local regression technique that describes spatial relationships between dependent and independent variables (Wang et al., 2013). The GWR variogram is mainly calculated through local regression by averaging over all pairs of observations (Fotheringham et al., 2003). The GWR modelling helps investigate the spatially nonstationary correlations among metals and the soil system's influencing factors in a cropping system. The results reflect these relationships and the acceptable threshold for the concentration of metals in a cropping system (Qu et al., 2014). In the current study, the GWR modelling approach was applied to evaluate correlations between the dependent variable (rice grain Cr) and independent variables of soil properties and the Cr in the paddy tissues (soil Cr, SOM, pH, root Cr, and shoot Cr). The GWR modelling approach has the critical ability to take into account the samples' spatial (three-dimensional) locations; this allows the variables to differ spatially and better reflects the differing correlations among the measured selected dependent and independent variables (Qu et al., 2015). The local regression parameters, i.e., the intercept, slope, and local R^2, were obtained through GWR modelling. The local R^2 is a measure of goodness of fit; its value ranges from 0.0 to 1.0, with higher values indicating a better fit (Comber et al., 2018). Local R^2 illustrates the level of variance in the dependent variable described through the regression model. The denominator for the R^2 calculation is the sum of the squared selected dependent variable values (Fotheringham et al., 2003). Therefore, the GWR model with measured selected dependent and independent variables was developed as follows (Fotheringham et al., 2003; Qu et al., 2015):

$$Y(u) = \beta_0(u) + \beta_1(\mathrm{u})\chi_1(u) + \beta_2(\mathrm{u})\chi_2(u) + \beta_3(\mathrm{u})\chi_3(u) + \beta_4(\mathrm{u})\chi_4(u) + \beta_5(\mathrm{u})\chi_5(u) + \varepsilon(u) \tag{3}$$

where Y(u) represents measured selected dependent variable paddy (grain Cr) at location vector (u); χ_1(u), χ_2(u), χ_3(u), χ_4(u), and χ_5(u) denoted measured selected independent variables (i.e., SOM, pH, soil Cr root Cr, and shoot Cr) at u, ε(u) is Gaussian error term at u, and β_0(u), β_1(u), β_2(u), β_3(u), β_4(u), and β_5(u) are the model intercept and the local regression coefficient (i.e., slopes) of the measured selected independent variables at u. the regression constants at u are assessed by a weighting function as follows (Fotheringham et al., 2003; Qu et al., 2015):

$$\widehat{\beta}(u) = \left[\mathrm{X^T W}(u) X\right]^{-1} \mathrm{X^T W}(u) Y \tag{4}$$

where X and Y denote simplistic arrays created in utilizing x and y variables, respectively; W(u) represents the local weight matrix, computed by a kernel function that places additional weight on the specific locations near calibration locations. The sample extents differed across sites; therefore, an adaptive bi-square kernel function was implemented for weight determination. The adaptive bi-square kernel function assigns a weight of zero to observations outside of the bandwidth, invalidating their influence on the local regression estimate (Bidanset and Lombard, 2014).

In this study, assessing the spatial nonstationarity is imperative to evaluate whether parameter approximations in the GWR assay significantly vary across the spatial area. The variability of the different variables' coefficients, the GWR model fitted by local data (usual GWR) with the different variables' coefficients being constant (mixed GWR) (Qu et al., 2014). We assessed that the usual GWR model is better than the mixed GWR model; as judged by a model comparison criterion such as the Akaike information criterion (AIC), we estimate that the different variable coefficients vary spatially. Therefore, in the current study, the best adaptive bandwidth for the model was selected based on the AIC to determine the optimal adaptive bandwidth of each model, which is a performance measure applied to compare models; a smaller AIC indicates a superior model (Li et al., 2010). Finally, 500 collected sample data points were employed to calibrate the GWR model at every specific spatial location; the AIC was calculated as follows:

$$\mathrm{AIC} = 2k - 2\mathrm{In}(L) \tag{5}$$

where k denotes the number of variables and L is the probability. The model with the lowest possible AIC presents the most effective modelling implementation and simplicity (Podgorski et al., 2017). The GWR 4.0 modelling program was applied in this study to perform the GWR analysis (Nakaya et al., 2014). ArcGIS 10.2 was utilized for the geostatistical analysis to produce maps, and OriginPro 8.5 was used to create graphs.

2.6. Chromium critical threshold in paddy soil systems

The CT for the concentration of paddy soil metals is defined as the maximum level of paddy soil metals that ensures that the level of metal of the rice gain is below its acceptable threshold (Qu et al., 2015). Thus, the CT index can show the rice grain sensitivity to specific metals in paddy soil systems. The CT values of paddy soil metal concentration at a particular sampling location are deduced using the allowable metal limit for rice grains and the local relationship between metal concentrations in rice grain and paddy soil and plant tissue influencing variables, which are calibrated through the GWR model.

In this study, the CT values of paddy soil Cr concentration at a specific sampling location were deduced using the allowable limit for rice grain Cr (1.0 mg kg^{-1}) and the local relationship between rice grain Cr concentrations, paddy soil (pH, SOM, and paddy soil Cr), and plant tissue variables (shoot Cr and root Cr), which were calibrated through the GWR model. The CT for the paddy soil Cr at every location was assessed as follows:

$$T_{soil\_Cr}(u) = \frac{P_{rice\_Cr} - \beta^0(u) - \beta_{pH}(u)X_{pH}(u) - \beta_{SOM}(u)X_{SOM}(u) - \beta_{root}(u)X_{root\_Cr} - \beta_{shoot}(u)X_{shoot\_Cr}}{\beta_{soil\_Cr}(u)} \tag{6}$$

where T_{soil_Cr} represents the CT for paddy soil Cr at each location (u), P_{rice_Cr} represents the allowable threshold for the rice grain Cr concentration (1.0 mg kg^{-1}), and X signifies the assessed value of the independent variable. The other symbols have the same meanings given in Eq. (3).

2.7. Chromium loading capacity in the paddy soil systems

The LC is defined as the variance between the CT and the actual metal concentrations in the soil system. The LC for the metal in the soil system indicates the contamination risk to a cropping system from the metal concentration in the soil system. In the current research, the LC values of paddy soil Cr are depicted as the difference between CT and actual paddy soil Cr concentrations (Qu et al., 2015). This LC index mainly represents the degree of risk of paddy soil Cr to rice grain. The LC at each assessed location (u) was measured as follows:

$$\widehat{LC}_{Soil\_Cr}(u) = \widehat{T}_{Soil\_Cr}(u)\widehat{X}_{Soil\_Cr}(u) \tag{7}$$

where $\widehat{T}_{Soil\_Cr}(u)$ and $\widehat{X}_{Soil\_Cr}(u)$ represent the valuations towards CT and the overall level of paddy soil Cr at each assessed location (u), respectively. A lower value of LC suggests a greater risk of paddy soil Cr concentrations being higher than the CT at each assessed location (u) and shows that the rice grain is at high risk from paddy soil Cr. The negative LC value for paddy soil Cr at each specific location (u) shows that the rice grain Cr concentration at location (u) is likely beyond the permissible threshold. Based on LC values, the risk areas were categorized as low-risk grade I (LC > 50 mg kg^{-1}), moderate-risk grade II (20 mg kg^{-1} ≤ LC ≤ 50 mg kg^{-1}), high-risk grade III (0 mg kg^{-1} ≤ LC < 20 mg kg^{-1}), and very high-risk grade IV (LC < 0 mg kg^{-1}).

2.8. Human health risk assessment index

The human health risk assessment index was linked to rice consumption by calculating the average daily dose (ADD, mg Cr kg^{-1} bodyweight ∗ day) for two target groups of adults and children, as follows (Rinklebe et al., 2019):

$$\mathrm{ADD} + \frac{\mathrm{Cr} \times \mathrm{ADC} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{8}$$

where Cr is the Cr (mg kg^{-1}) concentration in rice grain; ADC is the average daily (rice) consumption rate (kg day^{-1}); EF is the exposure frequency; ED is the exposure duration; BW is the average body weight; AT is the averaging time; This research obtained the ADC value in adults and children from a survey performed from 2015 to 2016 in Pakistan (Majeed et al., 2018).

2.8.1. Total hazard quotient (THQ)

THQ is the ratio of the exposure amount to the amount at which no toxic effects are likely to occur, even in sensitive individuals. The THQ for the consumption of contaminated rice can be estimated based on an oral reference dose (Rfd) (Li et al., 2014). The THQ (unitless) was calculated as follows:

$$\mathrm{THQ} = \frac{\mathrm{ADD}}{\mathrm{Rfd}} \tag{9}$$

The Rfd value for total Cr has not been defined yet; therefore, in this study, the Cr(III) Rfd value (1.5 mg kg^{-1} day^{-1}) was used for Cr, which was obtained from the US Environmental Protection Agency Regional Screening Levels (RSLs) (USEPA). Thus, a THQ $^{<}$ 1 indicates no risk of toxic, noncarcinogenic human health impacts, whereas a THQ $^{>}$ 1 indicates a high probability of adverse, noncarcinogenic human health effects (Li et al., 2014; Rinklebe et al., 2019). Further information about each value used for the variables in human health risk indices ADD and THQ calculation are summarized in Table S1.

3. Results and discussion

3.1. Physicochemical properties and chromium contamination of paddy soil

Paddy soil physicochemical variables (pH, SOM, and paddy soil Cr) used in the current study are displayed (Fig. 2 and S3) and summarized in Table 1. The overall average pH was 8.0 ± 0.4; in the study areas of Sindh and Punjab, the pH was 7.8 ± 0.3 and 8.1 ± 0.3, respectively. Overall, the average SOM concentration was 14.48 ± 5.76 mg kg^{-1}; the SOM concentrations in Sindh and Punjab were 11.61 ± 3.94 and 15.08 ± 5.89 mg kg^{-1}, respectively. The overall Cr concentration in paddy soil ranged from 61.82 to 173.34 mg kg^{-1}, with an average of 96.62 ± 16.59 mg kg^{-1}; with 62.6% samples exceeding the China natural background threshold value for Cr concentration in paddy soil 90 mg kg^{-1},

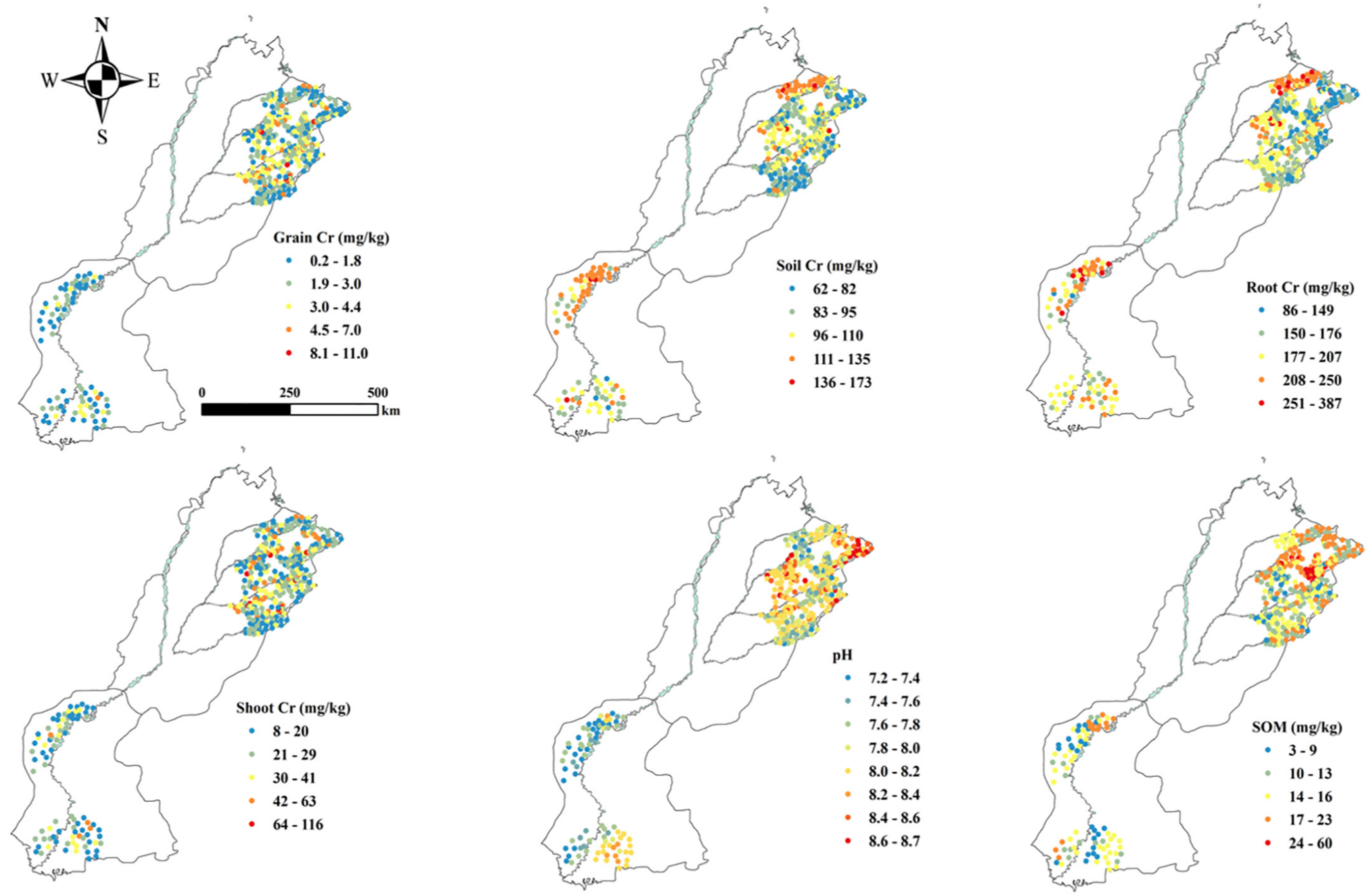

Fig. 2. Spatial maps show the distribution of Cr concentration and all measured parameters of paddy soil rice system samples collected from the Sindh and Punjab Provinces of Pakistan.

and all paddy soil samples are lower than the (pH-dependant > 7.5 threshold value for Cr 350 mg kg^{-1}, GB15618-2018). In the study areas of Sindh and Punjab, the Cr level in paddy soil ranged from 76.46 to 140.57 and from 61.82 to 173.34 mg kg^{-1}, with averages of 105.68 ± 14.57 mg kg^{-1} and 94.74 ± 16.38 mg kg^{-1}, respectively. In the study areas of Sindh and Punjab, 86.04% and 57.72% of the paddy soil samples exceeded the natural background threshold value of Cr 90 mg kg^{-1}. Notably, Pakistan does not have national regulatory limits for Cr in paddy soil or rice plants. Therefore, this study estimated risk by comparison to a separate country's standards (CNFS and EQSs). The average Cr concentration in paddy soil was comparatively higher than the levels found in several studies conducted in different parts of the world, including Shengzou (29.8 mg kg^{-1}) (Zhao et al., 2009), Kunming (73.4 mg kg^{-1}) (Cheng et al., 2018), Shimen, Fenghuang, and Xiangtan (26.0, 23.7, and 33.1 mg kg^{-1}, respectively) (Zeng et al., 2015), Jiaxing (60.63 mg kg^{-1}) (Qu et al., 2015), China; Faridpur, Bangladesh (86.0 mg kg^{-1}) (Ahsan et al., 2009); Narora, India (8.1 mg kg^{-1}) (Singh et al., 2014); and Aomori, Japan (31.0 mg kg^{-1}) (Tsukada et al., 2007).

3.2. Chromium paddy rice tissues and bioaccumulation and translocation factors

Chromium in paddy plant tissue concentrations in root samples from Sindh and Punjab ranged from 139.64 to 279.57 mg kg^{-1} and from 85.84 to 387.42 mg kg^{-1}, with averages of 202.45 ± 30.95 mg kg^{-1} and 177.65 ± 35.5 mg kg^{-1}, respectively. Those in shoots ranged from 8.20 to 55.11 mg kg^{-1} and 8.59 to 115.69 mg kg^{-1}, with averages of 22.47 ± 9.42 mg kg^{-1} and 28.13 ± 14.05 mg kg^{-1}, respectively. The overall Cr in rice grain ranged from 0.20 to 10.99 mg kg^{-1}, with an average of 2.68 ± 1.46 mg kg^{-1}. In the study areas of Sindh and Punjab, these values ranged from 0.56 to 5.05 mg kg^{-1} and from 0.20 to 10.99 mg kg^{-1}, with averages of 1.96 ± 1.00 mg kg^{-1} and 2.82 ± 1.50 mg kg^{-1}, respectively (Fig. 2, S3, and Table 1). In Sindh and Punjab, 90.69% and

Table 1
Statistical summary of Cr concentration and all measured parameters of paddy soil-rice system samples collected from Sindh and Punjab Provinces of Pakistan.

Variables	Permissible limit	Sindh (n = 86)			Punjab (n = 414)		
		Minimum	Maximum	Average ± Std. deviation	Minimum	Maximum	Average ± Std. deviation
Grain Cr (mg kg^{-1})	1.0 (CNFS)	0.56	5.05	1.96 ± 1.00	0.20	10.99	2.82 ± 1.50
Shoot (mg kg^{-1})	–	8.20	55.11	22.47 ± 9.42	8.59	115.69	28.13 ± 14.05
Root (mg kg^{-1})	–	139.64	279.57	202.45 ± 30.95	85.84	387.42	177.65 ± 35.55
Soil (mg kg^{-1})	90 (EQSs)	76.46	140.57	105.68 ± 14.57	61.82	173.34	94.74 ± 16.38
pH	7–8.5	7.2	8.4	7.8 ± 0.3	7.2	8.7	8.1 ± 0.3
SOM (mg kg^{-1})	–	6.02	18.69	11.61 ± 3.94	3.24	59.50	15.08 ± 5.89

Number of samples (n), natural background threshold value regulates by China Environmental Quality Standards (EQSs)l, and China National Food Standard (CNFS).

98.79%, respectively, of grain samples exceeded the allowable threshold for Cr in grain (1.0 mg kg^{-1}) established by the CNFS. Similarly, the average soil Cr and the average Cr concentration in rice grains are comparatively higher than those reported in several studies in different parts of the world, including Shengzou (0.2 mg kg^{-1}) (Zhao et al., 2009), Shimen and Fenghuang (0.1 mg kg^{-1}) (Zeng et al., 2015), Jiaxing (0.46 mg kg^{-1}) (Qu et al., 2015), China, and Narora, India (0.6 mg kg^{-1}) (Singh et al., 2014).

Similarly, regarding the Cr concentration in paddy tissues, rice roots have the highest Cr BAFs. In the study area of Sindh, root BAFs ranged from 1.22 to 2.62, with averages of 1.93 ± 0.22, shoots ranged from 0.07 to 0.63, with averages of 0.22 ± 0.1, and grains ranged from 0.001 to 0.006, with averages of 0.002 ± 0.01. In the study area of Punjab, roots ranged from 0.03 to 2.85, with averages of 1.89 ± 0.27, shoots ranged from 0.08 to 1.38, with averages of 0.31 ± 0.17, and grains ranged from 0.02 to 0.13, with averages of 0.03 ± 0.02 (Table S2). Consequently, in the study area of Sindh, TF in aerial tissue shoots ranged from 0.04 to 0.28, with averages of 0.12 ± 0.05, and grain ranged from 0.003 to 0.03, with averages of 0.01 ± 0.01. In the study area of Punjab, shoots ranged from 0.04 to 1.04, with averages of 0.16 ± 0.1, and grain ranged from 0.001 to 0.1, with standards of 0.02 ± 0.01.

Chromium levels in paddy plant aerial tissues (shoot and rice grain) significantly depend on the availability of soil Cr and the rice uptake ability (Infante et al., 2021). In the current study, as Fig. 4S shows, the Sindh and Punjab samples had BAFs $^{>}$ 1, which indicated that Cr significantly accumulated in paddy root tissues (Fig. 4S). Roots are the primary tissues that take up Cr for rice plants; the highest bioaccumulation capacity of Cr in roots is significantly ascribed to its unique role in adsorbing, retaining, and translocating Cr from paddy soil to roots. In this study, Cr BFs and TF from root to above-ground paddy tissues (shoot and rice grain) were low, which suggested that Cr is not well translocated and accumulated in the shoots and rice grain and instead concentrated in the plant's root cells. Different studies have indicated that Cr immobilization mainly occurs in the vacuoles of root cortex cells; thus, the accumulation of Cr in paddy root tissues is high (Hayat et al., 2012; Zeng et al., 2010). Furthermore, highly mobile Cr(VI) can be reduced to less mobile Cr(III) and stored in vacuoles of root cortex cells as a likely natural toxicity response of paddy plants (Infante et al., 2021).

To describe the influence of the paddy soil and plant tissue variables (SOM, pH soil Cr, root Cr, and shoot Cr) on Cr accumulation in rice grains, we computed Pearson's nonparametric correlation coefficients among paddy soil and plant tissue variables and rice grain Cr (Table S3). The results suggest that rice grain Cr is positively correlated with the shoot Cr concentration (correlation coefficient $r = 0.89$) and weakly correlated with soil Cr ($r = 0.12$). The root Cr concentration was positively correlated with the paddy soil Cr concentration ($r = 0.74$) and weakly correlated with the shoot Cr concentration ($r = 0.09$), grain Cr level ($r = 0.12$), pH ($r = 0.160$), and SOM ($r = 0.08$); all these correlations were considered significant at the 0.05 level. These correlations indicated that soil Cr and soil pH play an important role in Cr accumulation in paddy roots, shoots, and rice grains. The shoot Cr was the most significant factor among all the considered factors because it had a high correlation coefficient with the rice grain Cr. However, the correlation coefficient between soil and plant tissue variables was not significant at the 0.01 level. Thus, this study adopted GWR models of rice grain Cr on soil and plant tissue variables (i.e., pH, SOM, soil Cr, root Cr, and shoot Cr).

3.3. Elucidating spatially nonstationary correlations between selected dependent and independent variables

The variogram GWR modelling results; the local regression parameters for the 500 data points in the study area, i.e., the intercept, slope, and local sum of the squared dependent variable values (R^2) obtained through GWR modelling of the dependent variable (rice grain Cr) concentrations against the independent paddy soil (pH, SOM, and paddy soil Cr) and plant tissue variables (shoot Cr and root Cr); and their spatial distribution in the study area of Sindh and Punjab are shown in Fig. 3 and summarized in Table S4. The regression coefficient of these relationships was applied to elucidate the combined influences of independent variables on Cr accumulation in grains. In the estimation, the GWR mainly moves a weighted window over the spatial data and estimates the impartial slope and model intercept (mean value of response variable in predictor variables in the model) from the fit point and estimated dependent variable compared from a distance matrix of all points within the data set (Wheeler and Páez, 2010). Whereas in conventional linear regression analysis and ordinary least squares regression method, which can estimate the intercept regression coefficient based on the assumption of observations being independent of each other, which fails to evaluate the model intercept spatial dependence of the variables over varied space (Qu et al., 2014). In the study area of Sindh, the model intercept measured Cr rice grain-dependent variables related to independent soil properties, and plant tissue variables ranged from −0.872 to 1.157, with an average of −0.108 ± 0.649. The regression coefficient pH and SOM ranged from 0.053 to 0.279 and −0.009 to −0.003, with averages of 0.191 ± 0.073 and −0.007 ± 0.001, respectively. Whereas the regression coefficient soil Cr ranged from −0.014 to −0.008, with an average of −0.010 ± 0.002, the regression coefficient root Cr ranged from −0.001 to 0.00001, with an average of −0.001 ± 0.0005. No variation was observed in the regression coefficient of the measured shoot Cr value. At the same time, the local R^2 values ranged from 0.702 to 0.721, with an average of 0.710 ± 0.006.

Likewise, in the study area of Punjab, the model intercepts ranged from −3.256 to 3.218, with an average of 0.412 ± 0.1300. The pH and SOM regression coefficients ranged from −0.400 to 0.414 and −0.035 to 0.012, with averages of −0.016 ± 0.169 and −0.009 ± 0.010, respectively. Whereas the regression coefficient of soil Cr ranged from −0.006 to 0.004, with an average of −0.002 ± 0.003, the regression coefficient of root Cr ranged from −0.001 to 0.004, with an average of 0.001 ± 0.001. The regression coefficient of shoot Cr ranged from −0.090 to 0.098, with an average of 0.094 ± 0.002, and the R^2 ranged from 0.717 to 0.876, with an average of 0.813 ± 0.048. In both Sindh and Punjab Provinces of Pakistan study areas, the overall AIC = 1776.746 and the sum of squared residuals (SSR) = 79.868 were measured for the rice grain Cr dependent variable.

These diverse spatial regression parameters confirmed that the analysis was not constant across the study area. The relationships between dependent and independent variables were spatially nonstationary relationships that varied from one location to another across the research area. A positive coefficient value indicates positive effects, while a negative coefficient value suggests a negative effect (Qu et al., 2014). Likewise, an exceptionally high absolute regression coefficient value signifies a strong relationship. In contrast, a low total regression coefficient value suggests a weak relationship between variables (Qu et al., 2015). The GWR modelling results indicate that the strength of the influence-independent variables on Cr accumulation in rice grains varies from one location to another throughout the research area, as indicated by the differences in regression coefficient values. In the paddy soil system, most trace elements, including Cr, are bound to minerals and passive SOM and thus are not phytoavailable (Antoniadis et al., 2017; Shahid et al., 2017). Although active SOM can increase the phytoavailability of Cr by increasing the CEC of the paddy soil system due to its role as an electron shuttle for the bioreduction process, it can significantly increase the chelation and retention of Cr and enhance nutrient solubility in the soil solution (Jardine et al., 2013; Shahid et al., 2017). Therefore, the effect of SOM on Cr bioavailability and accumulation in the plant system depends on its concentration and its characteristics. Likewise, the phytoavailability of Cr in plant systems varies with the soil pH and decreases when pH < 7 causes desorption of Cr; in contrast, at pH > 8, Cr precipitates inside the soil matrix. In this way, Cr has solubility, bioavailability, and mobility at lower pH values (Ma and Hooda, 2010; Shahid et al., 2017). At decreased/low soil pH values, H^+ competes for the available binding sites and increases Cr release to the soil solution from the soil binding site. In contrast, the absence of protons under high pH

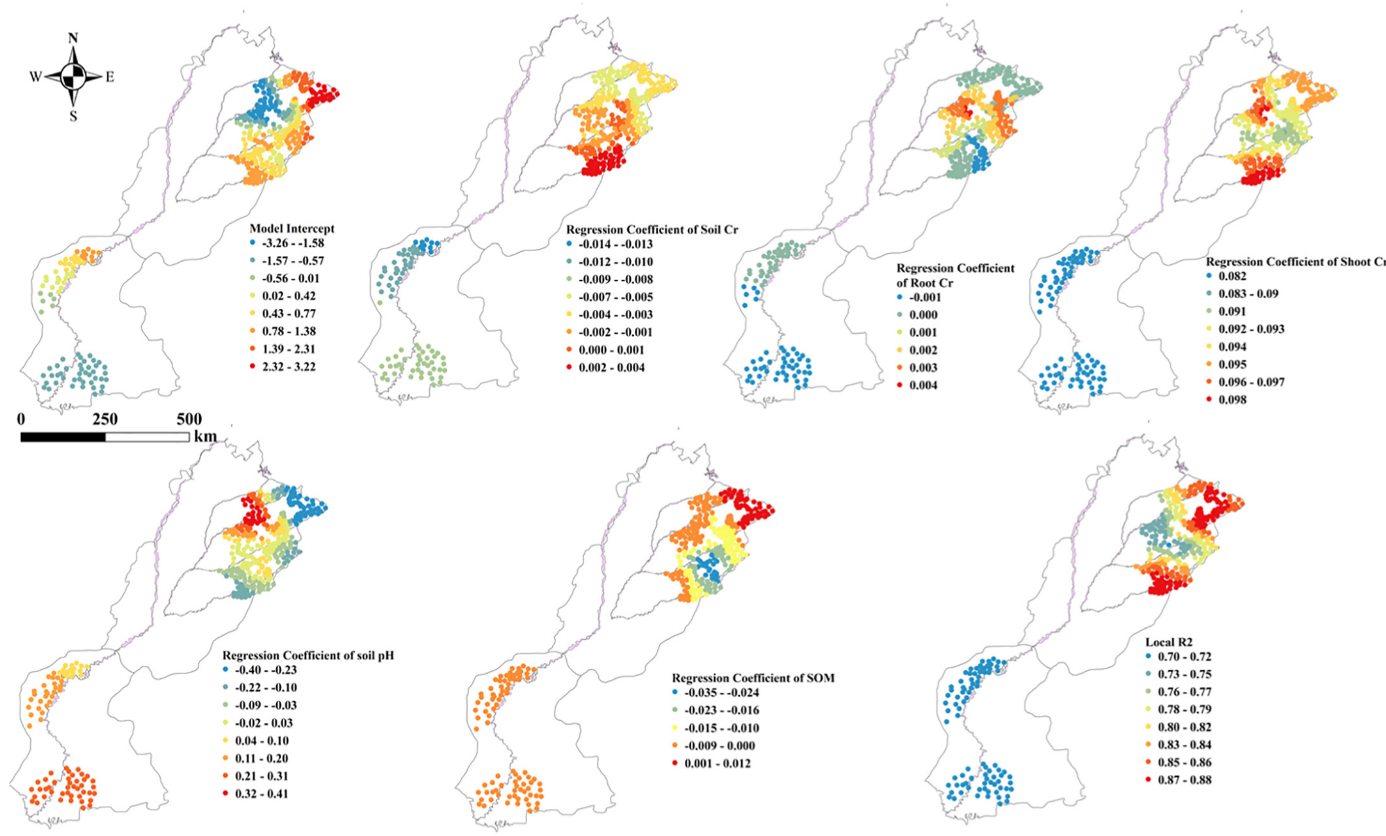

Fig. 3. Geographically weighted regression (GWR) modelling variable analysis at sampling locations in the Sindh and Punjab Provinces of Pakistan for rice grain Cr against plant tissues (root and shoot) and paddy soil properties (model intercept regression coefficient of soil pH, regression coefficient of root Cr, regression coefficient of shoot Cr, regression coefficient of soil Cr, local R^2, and regression coefficient of soil organic matter (SOM)).

conditions provides solid phase-exchange sites for Cr or other trace element cations (Zeng et al., 2011).

Therefore, the combined effects of the pH, SOM, and soil Cr on Cr phytoavailability and Cr accumulation in the paddy plant system varied across the study area. The reasons mentioned above likely explain why the effects of soil properties on the phytoavailability and accumulation of Cr in the paddy plant system varied spatially over the study area (i.e., they showed evident spatially nonstationary relationships between dependent and independent variables). The other factors, such as CEC, E_h, clay minerals contents, and Fe/Mn oxides, which were not selected or unavailable in this study, may also influence Cr accumulation in the paddy plant system. If they have an evident impact, the spatial dissimilarities of these hidden factors may also change relationships between Cr in the rice system and the selected factors across the study area. The paddy rice cultivar may also significantly influence Cr accumulation in paddy systems (Qu et al., 2015).

Several studies have indicated that, compared with conventional rice, hybrid rice has a greater uptake, transport, and bioaccumulation capability of trace metals, including Cr, in paddy tissue roots, shoots, and rice grains (Xie et al., 2016; Ye et al., 2012). In the current study, hybrid paddy rice cultivars were dominant in Sindh and conventional basmati in Punjab (Khushik et al., 2011). Therefore, basmati or other hybrid paddy rice varieties had been cultivated in the research area, and the influence of the associated paddy soil variables on Cr phytoavailability and accumulation in rice would not be precisely the same (Cao et al., 2014; Norton et al., 2009; Qu et al., 2015; Zeng et al., 2008). Consequently, the Cr contamination risk in paddy soil systems is directly linked to the variety of cultivated paddy rice.

The independent explanatory variables and the power of the GWR model varied across space, as confirmed through the regional values of the R^2 for the model shown in Fig. 3. Considering the redundancy between independent variables such as pH, SOM, soil Cr, root Cr, and shoot Cr, their combined influence on the accumulation of Cr in grains was explored. Fig. 3 shows that these independent critical factors could elucidate Cr accumulation in paddy rice grains. Hence, adaptive bandwidth in GWR modelling varied across the study area with the regional concentration of sampling points. As bandwidth increases or decreases, the GWR modelling assessment becomes increasingly regional or global, revealing topographical aspects such as a "spatial microscope" (Fotheringham et al., 2003; Qu et al., 2015). The scarcity of sampling points in the central regions of the study areas of Sindh and Punjab Provinces may provide a useful example of an instance in which the GWR model's explanatory power is comparatively weak. Therefore, to overcome this limitation of the GWR model's weak explanatory power was fixed in these areas by simply applying adaptive kernel density estimation relative to the adaptive kernel's points with a non-zero weight (DesMarias and Costa, 2019; Wheeler and Páez, 2010). Consequently, the local R^2 regression observations continually cross the areas where the GWR model's explanatory power is weak.

3.4. Spatially critical threshold and loading capacity distribution and elucidating risk areas for paddy soil chromium accumulation in rice grains

The CT values for the paddy soil Cr across the study areas of Sindh and Punjab sampling locations were deduced using the allowable rice Cr threshold from the CNFS standard (1.0 mg kg^{-1}). The relations among dependent (rice grain Cr) and independent paddy soil (pH, SOM, and paddy soil Cr) and plant tissue variables (shoot Cr and root Cr) were calibrated with the GWR model approach. Throughout the study area, the overall CT value in the paddy soil system ranged from 50.03 to 301.97 mg kg^{-1}, with an average of 124.97 ± 61.81 mg kg^{-1}, and a total of 38.4% of the paddy soil samples exceeded the CT values, indicating the risk of Cr in paddy soil (Table S5). In the study area of Sindh paddy soils, the CT values ranging from 151.92 to 301.97 mg kg^{-1} with an average of 215.12 ± 35.57 mg kg^{-1} were higher than the measured Cr concentrations ranging from 76.46 to 140.57 mg kg^{-1} with an average of 105.68 ± 14.57 mg kg^{-1}; these results revealed that the risk of paddy soil Cr exceeding the CT value was relatively low in this area. In the study area of Punjab paddy soils, the CT values ranged from 50.03 to 293.14 mg kg^{-1} with an average of 106.25 ± 48.10 mg kg^{-1}, and 46.4% of samples were lower than the measured Cr concentrations ranging from 61.82 to 173.34 mg kg^{-1} with an average of 94.74 ± 16.38 mg kg^{-1}, suggesting that the risk of paddy soil Cr exceeding the CT value is relatively high in this area. The spatial distribution map of the CT of the paddy soil Cr is shown in Fig. 4a. Low CT values occur in the northeastern, southeastern, and central parts of the Punjab research area. These low CT values for Cr in paddy soil suggest that Cr in rice grains in these research areas is critically sensitive to paddy soil Cr. In the study area of Sindh, the CT values for Cr in soil were high, indicating that rice in this part of the study area is not as sensitive to paddy soil Cr as that in the study area of Punjab Province as compared to TF of Cr to grains (Fig. S5).

The LC concept reflects the degree of risk of paddy soil Cr accumulation in rice grains. Throughout the study area, the overall LC value in the paddy soil system ranged from – 97.11 to 161.49 mg kg^{-1}, with an average of 28.35 ± 56.52 mg kg^{-1}. In the study area of Sindh, the LC values ranged from 73.50 to 161.40 mg kg^{-1} with an average of 109.43 ± 21.86 mg kg^{-1}. In the study area of Punjab, the LC values ranged from – 97.11 to 161.49 mg $kg^{-1,}$ with an average of 11.51 ± 45.92 mg kg^{-1} (Table S6). The overall LC spatial distribution tendency of the differences between the CT and the measured LC values was not substantially different from the paddy soil Cr distribution and the CT values throughout the study area. The spatial distribution map of the LC values for the paddy soil Cr is analogous to the spatial distribution pattern of the CT of the paddy soil Cr, as shown in Fig. 4b. The areas with identified LC values of paddy soil Cr were negative, suggesting that rice grain Cr may have already exceeded the CNFS allowable threshold (1.0 mg kg^{-1}).

The spatial distribution map of risk areas is mainly generated as LC (mg kg^{-1}) maps based on paddy soil Cr risk grades: low-risk grade I (LC > 50), moderate-risk grade II (20 ≤ LC ≤ 50), high-risk grade III (0 ≤ LC < 20), and very high-risk grade IV (LC < 0). Throughout the study area, based on the LC% of the samples, the overall paddy soil Cr risk areas with grade I (34.6%), II (15.8%), III (11.2%), and IV (38.4%) are summarized in Table S6. In the study area of Sindh, grade I (100%) paddy soil Cr risk occurred in the entire study area where the highest LC values were observed (Fig. 4c). Whereas in the study area of Punjab, the grade I and II (21.0% and 19.1%, respectively), paddy soil Cr risk areas mainly occurred in the central part of the northwestern and some parts of the southeastern regions. The grade III (13.5%) paddy soil Cr risk areas occurred in the upper and central parts of the northeastern and some lower parts of the southeastern regions, where lower LC values were observed for the paddy soil Cr. In contrast, in the study area of Punjab, grade IV (46.4%) paddy soil Cr risk areas mainly occurred in the central parts of the northeastern areas, where negative LC values were observed.

Thus, intensive monitoring is highly recommended for grades III and IV paddy soil Cr risk areas to know key Cr accumulation sources in the paddy system. After that, remediation strategies might be considered next if feasible. This extensive study in Sindh and Punjab Provinces of Pakistan rice-growing regions assessed contamination risk from paddy soil Cr based on a random, nonspecific sampling of different rice cultivar varieties. However, the contamination risk of paddies to different paddy rice varieties might vary because the capability of paddy crops to absorb or accumulate metals might differ among rice cultivars. Hence, further paddy rice variety-based studies are recommended, particularly in these rice-growing regions of Pakistan.

3.5. Health risk assessment of chromium exposure through rice consumption

Humans' exposure to metals involves several pathways, such as dermal contact, drinking water, soil ingestion, and inhalation routes (Guo et al., 2020). At the same time, dietary consumption has been identified as a critical pathway contributing to approximately 90% of human health risks (Doabi et al., 2018; Zhang et al., 2020). Consequently, this study focused on Cr-associated human health risks through reported average daily rice consumption by adults and children (108 and 56.6 g rice day^{-1}, respectively) (Majeed et al., 2018). The health risk assessment indices ADD and THQ for adults and children in the study areas of Sindh and Punjab Provinces of Pakistan are given (Table S7). The ADD (mg Cr kg^{-1} bodyweight ∗ day) values for Cr for adults in Sindh and Punjab ranged from 0.0004 to 0.003 with an average of 0.001 and from 0.0001 to 0.01 with an average

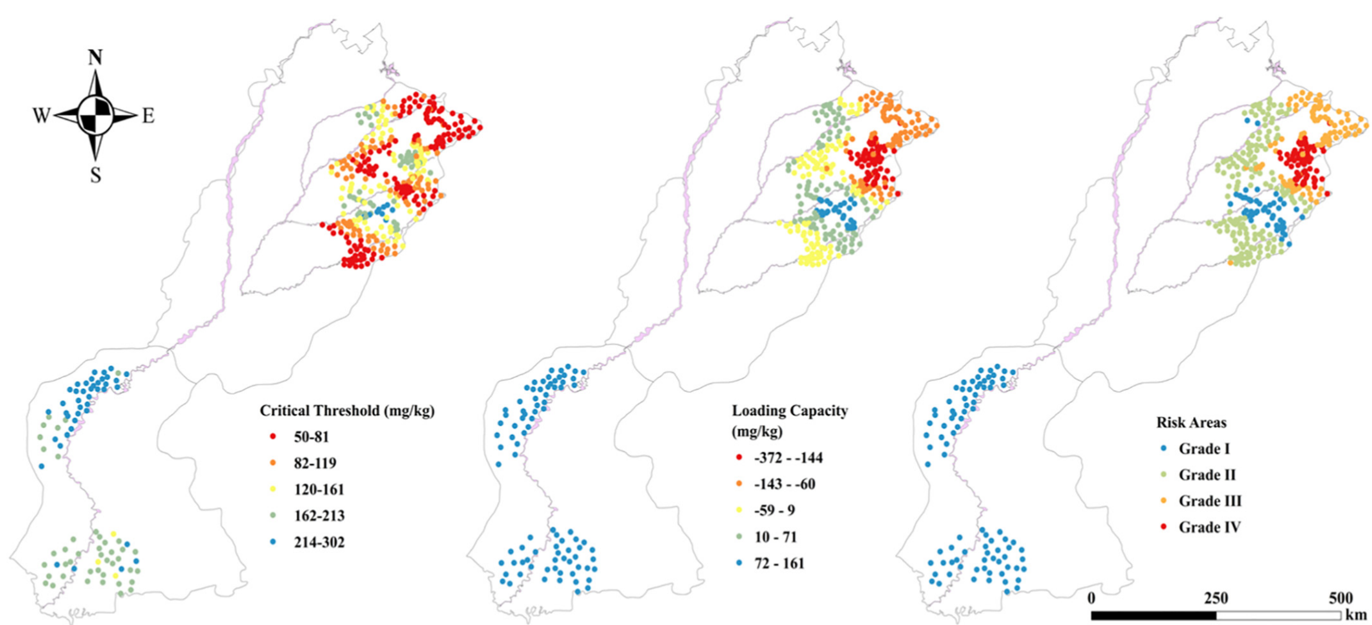

Fig. 4. Spatial maps show the distribution of the critical threshold (CT) of Cr in paddy soil that may confirm that the concentration of grain Cr is lower than the allowable threshold of grain Cr (a), the loading capacity (LC) of Cr in paddy soil at the sampling location (b), and the risk areas at the sampling locations, Sindh, and Punjab Provinces of Pakistan. Risk areas are defined as at low-risk grade I (LC > 50 mg kg^{-1}), moderate-risk grade II (20 mg kg^{-1} ≤ LC ≤ 50 mg kg^{-1}), high-risk grade III (0 mg kg^{-1} ≤ LC < 20 mg kg^{-1}), and very high-risk grade IV (LC < 0 mg kg^{-1}) (c).

of 0.002, respectively, and those for children ranged from 0.002 to 0.01 with an average of 0.01 and from 0.001 to 0.03 with an average of 0.01, respectively. The THQ values in Sindh and Punjab for adults ranged from 0.0002 to 0.002 with an average of 0.001 and from 0.0001 to 0.01 with an average of 0.001, respectively, and those in children ranged from 0.001 to 0.01 with an average of 0.04 and from 0.0004 to 0.02 with an average of 0.005, respectively. The results suggested that the health risk indexes (ADD and THQ) based on rice consumption in adults and children in the study area of Punjab are relatively higher than those in the study area of Sindh.

A study conducted in the Jin Qu Basin of China reported the average Cr ADD (1.5) and THQ (0.001), where the average rice consumption rate is much higher (238 g kg^{-1} day^{-1}) (Guo et al., 2020) than that in Pakistan. In this study, the measured average THQ (0.001), consistent with the above research values, among samples from Sindh and Punjab, Provinces, which had THQ values $\ll 1$, indicates no potential health risk originating from Cr exposure. Although the health risk indexes ADD and THQ show no potential health risk, 97.4% of grain samples exceeded the Cr threshold values 1.0 mg kg^{-1}, as determined by CNFS, Cr concentration in paddy soil, 62.6% samples exceeding the China natural background threshold value (90 mg kg^{-1}) as determined by China EQSs indicating they are still a concern.

To further reduce Cr contamination in the paddy soil-rice system, mitigation actions might be implemented, such as increasing soil pH to minimize the phytoavailability of soil Cr (Anemana et al., 2020; Scattolin et al., 2021) and planting other crops that exhibit a lower tendency for Cr uptake, transport, and accumulation (Tangahu et al., 2011) are mainly suggested for the high-risk zones in the study area. This study was carried out to assess Cr-associated human health risk assessment through rice consumption. However, exposure to Cr can occur through other sources, such as contaminated vegetables and fruits, wheat, dust inhalation, and dermal contact, and studies incorporating these additional sources of exposure are also recommended.

4. Conclusion and perspective

This detailed study found that 97.4% of rice grain samples exceeded the threshold value (1 mg kg^{-1}) determined by the CNFS. Overall Cr concentration in paddy soil, 62.6% samples exceeding the China natural background threshold value (90 mg kg^{-1}) for Cr concentration in paddy soil, and lower than the (pH-dependant > 7.5 standards for Cr 350 mg kg^{-1}, GB15618-2018) as determined by China EQSs. The GWR modelling investigation revealed that the spatial autocorrelations of residual relationships between the dependent (rice grain Cr) and independent paddy soil (pH, SOM, and paddy soil Cr) and plant tissue variables (shoot Cr and root Cr) on a regional scale in terms of global indices were not constant in the study area, representing great spatial nonstationarity.

The CT distribution findings show that 38.4% of the paddy soil samples exceed CT values, which confirms the high risk of paddy soil Cr in the study area and that rice grains are sensitive to paddy soil Cr. Additionally, the LC for Cr in the paddy soil was used to identify potential Cr contamination risk areas. Based on LC (mg kg^{-1}) values, the Cr risk within the study area was categorized as low-risk grade I (LC>), moderate-risk grade II ($20 \leq LC \leq 50$), high-risk grade III ($0 \leq LC < 20$), and very high-risk grade IV ($LC < 0$). In the study area of Punjab, LC % of samples low-risk grade I (21%) and moderate-risk grade II (19.1%), the paddy soil Cr risk areas occurred in the central part of the northwestern and southeastern regions. High-risk grade III (13.5%) paddy soil Cr risk areas occurred in the upper and central parts of the northeastern and lower parts of the southeastern areas, where lower LC values were observed. Very high-risk grade IV (38.4%) paddy soil Cr risk areas mainly occurred in the central parts of the northeastern Punjab study area, where negative LC values and samples exceeding CT values were observed. In the study area of Sindh, low grade I (100%) paddy soil Cr risk occurred in the entire study area where the highest LC values were observed.

The human health risk index THQ for samples from Sindh and Punjab had values $\ll 1$, indicating no potential health risk originating from Cr exposure for adults and children. Though the health risk index THQ results suggest no potential health risk, the paddy soil and rice grain concentration is high and exceeds the Cr threshold values. Thus, the government of Pakistan and associated environmental agencies may conduct further assessments pointing out the dynamics of sources of Cr pollution and deal with such contamination. Therefore, intensive monitoring could be the first step in this scenario, prioritizing the high- and very high-risk grades paddy soil Cr risk areas. Furthermore, this study will help fill the research gaps in other rice-growing regions.

CRediT authorship contribution statement

Waqar Ali: Conceptualization, Methodology, Software, Data curation, Writing - Original draft preparation. **Kang Mao**: Conceptualization, Methodology, Writing - Reviewing and Editing. **Muhammad Shafeeque**: Writing - Reviewing, and GIS mapping. **Muhammad Wajahat Aslam**: Sampling, Writing - Reviewing and Editing. **Xuefeng Yang**: Writing - Reviewing and Editing. **Li Zhong**: Writing - Reviewing and Editing. **Xinbin Feng**: Writing - Reviewing and Editing. **Hua Zhang** and **Joel Podgorski**: Supervision, Conceptualization, Methodology, Writing - Reviewing and Editing.

Declaration of competing interest

The authors declare no competing financial interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.153910.

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