



Research Article

Spatial Variability Assessment of Available Manganese Content in Soils of Jeedimetla Industrial Belt of Hyderabad, India

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ABSTRACT

The spatial distribution of heavy metals content in soils could help identify the hot-spot areas of contamination and potential sources of pollutants. A study was conducted at the Jeedimetla Industrial belt, located in the northwest part of Hyderabad, Telangana, to assess the spatial variability of soil DTPA extractable manganese (Mn) content. A total 100 number of surface (0-15cm depth) soil samples were collected from the study area, considering various land use like residential, industrial, agricultural, and scrub/wasteland and using a stratified random sampling method. The soil samples were analyzed using the standard analytical procedure for pH, electrical conductivity, oxidizable organic carbon content, and available Mn. The soil samples were split into calibration (75% of samples) and validation (25%) datasets for spatial prediction of Mn content using the geostatistical method, i.e., ordinary kriging. The spatial pattern of Mn was assessed using semivariogram analysis. Results show that the mean observed Mn content of calibration and validation datasets were 26.6 and 28.5 mg kg⁻¹ with a coefficient of variation of 0.66 and 0.55, respectively. The results revealed that the mean DTPA extractable Mn content of the study area exceeds the permissible limit of 10 mg kg⁻¹. The highest and lowest available Mn was detected in an agricultural land and residential area respectively. The Mn content shows moderate spatial dependency, which is explained better through a stable model (nugget to sill ratio of 0.51). The mean predicted Mn content was 26.5 mg kg⁻¹ at the study site. The results of the accuracy assessment show that the ordinary kriging with a stable variogram model could predict Mn content in an unsampled location with an accuracy of (coefficient of determination; R² of 0.61) and RMSE and MAD of 9.0 and 6.54 mg kg⁻¹, respectively. The results help to know the spatial distribution pattern of Mn content in the study area, which can help formulate the management plan to reduce the risk of soil pollution by Mn.

Key words: Geo-statistics, Heavy metals, Kriging, Semivariogram, Soil pollution

Introduction

Soil is one of the practical life support systems on Earth. The safety and quality of the soil

environment are crucial for maintaining continuous socioeconomic development and safeguarding human health. Due to the rapid rise of the global economy and urbanization, researchers have emphasized environmental problems related to soil heavy metal contamination (Chen *et al.*, 2019).

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Heavy metals are necessary to both plants and animals in trace amounts, such as copper (Cu), cadmium (Cd), zinc (Zn), manganese (Mn), cobalt (Co), and molybdenum (Mo), act as micronutrients for the growth of animals and plants, if the concentration exceeds the critical limit, it becomes toxic. (Tilwankar *et al.*, 2018). Manganese (Mn) is widely dispersed in soil, sediments, water, and biological components. It is the second most common transition metal in the Earth's crust. Free metal ions and soluble or insoluble metal complexes are a few forms of Mn (Geszvain *et al.*, 2012). Between 450 and 4000 mg kg⁻¹ of Mn is readily available in soils, and its availability can quickly rise when the pH of the soil drops or when the soil is reduced Sparrow and Uren (2014). Kaur and Rani (2006) and Singh *et al.* (2020) reported a <4, 4-10 and >10 ppm Mn content in soils as deficient, desirable and permissible limit. Mn is a heavy metal; at excessive farmlands levels, it reduces crop output and quality and poses a hazard to human health (Nagajyoti *et al.*, 2010). Mn accumulation in humans affects the central nervous system, resulting in Parkinson-like illnesses (Lucchini *et al.*, 2017). Excessive Mn in plants has adverse effects at different morphological scales, resulting in symptoms including chlorosis and necrosis, wrinkled leaves, brown patches, and growth inhibition.

Thus it is necessary to detect the concentration of Manganese in the soils. The basic approach to determining heavy metal (Mn) content is based on a traditional soil sampling design with a soil measurement depth of 0–15 cm and subsequent laboratory chemical analysis of the collected soils. However, this method is time-consuming and expensive to calculate the concentration of heavy metals in vast areas. Geostatistics offers an enhanced methodology that enables spatial interpolation across a broader area and makes it easier to quantify the spatial aspects of soil parameters. In recent years, geostatistics has been used to examine heavy metal soil pollution (Hani *et al.*, 2014; Wu *et al.*, 2009; Chandrasekaran *et al.*, 2015). This method used the stochastic theory of spatial correlation for interpolation and interpolation-related uncertainty. Geostatistics takes advantage of this knowledge. Geostatistics uses statistical variation understanding as a significant source of information to enhance the

estimation of an attribute at non-sampled locations with a sampled dataset.

Additionally, these techniques offer a collection of statistical instruments for including spatial coordinates of observations in data processing (Goovaerts, 1997). In order to compute and model the variograms and predict the concentration in non-sampled areas using kriging and statistical analysis of errors, geostatistical analysis treats the concentration of a potentially hazardous element in an affected medium as a regionalized variable in space. In order to examine environmental soil pollution, geostatistical analytic techniques like ordinary kriging, regression kriging, and indicator kriging have been employed extensively (Cheng *et al.*, 2013). By keeping the views mentioned above in mind, the present study was undertaken at the Jeedimetla industrial belt of Hyderabad to assess the spatial variability of soil Mn content using a geostatistical approach. Moreover, several studies were conducted at the experimental site mainly focused on water pollution (Piska *et al.*, 2004; Tilwankar *et al.*, 2018) by heavy metals rather than soil pollution study, which is scanty. Hence the findings of the present study will be a spatial inventory of Mn concentration in soils of the Jeedimetla Industrial belt.

Materials and Methods

Study area

Jeedimetla is situated (17.5197° N, 78.4586° E) in the Medchal-Malkajgiri district of the North-west part of Hyderabad, Telangana (Fig. 1). The total area of the study site is 13.6 km². The average sand, clay and silt content of Jeedimetla soils are 64.7, 19.9, and 15.4% respectively; hence the soils are sandy clay loam in nature. Jeedimetla experience a tropical climate, with a mean annual temperature and precipitation of 26.9°C and 855mm respectively. Jeedimetla is one of the industrial hub of Hyderabad, consisting of many industries like Pharmaceutical, Dyes, Steel, etc. Central Pollution Control Board (CPCB), reported the fact that the groundwater of Jeedimetla, as well as its adjoining areas, has arsenic, nickel, cadmium, and other heavy metals concentration more than the permitted level (Bhawan and Nagar, 2020).

Soil sampling and preparation

100 surface (0–15 cm depth) soil samples were collected from October 2021 to December 2021 from Jeedimetla using stratified random sampling (Fig. 1). The initial soil sampling plan was formulated using land use information derived from a land use and land cover map (1:50K scale) obtained from Bhuvan, NRSC, ISRO (bhuvan.nrsc.gov.in). However, based on accessibility, soil samples were collected from various land use, including industrial areas, residential areas, agricultural fields, and barren/wastelands over the study site. 26, 25, 20, and 29 sites were covered under industrial, residential, agricultural, and barren/wastelands, respectively. The soil samples were collected using a soil auger, placed in a polythene bag with a label describing the sampling details, and transported to the Professor Jayashankar Telangana State Agricultural University, Central Instrumental Cell, for additional processing and analysis. The sampling location's coordinates were recorded using a handheld Global Positioning System (GPS). The collected samples were brought to the laboratory and shade dried. The clods in the

sample are pounded using a wooden mortar and pestle and sieved the sample using a 2mm sieve for further analysis.

The soil samples were analysed for physicochemical properties like pH, Electrical Conductivity (EC), soil organic carbon (SOC), and available Mn content. The samples were divided into calibration and validation, which have 75% and 25%, respectively.

Laboratory analysis

The potentiometric approach was used to measure the pH of the soil suspension (Jackson, 1973), with the ratio of soil to distilled water suspension being 1: 2.5. A digital conductivity meter was used to measure EC in the suspension after it had been set aside for two hours (Jackson, 1973). The Rapid Titration method measured the amount of oxidizable SOC content (Walkley and Black, 1934). Manganese was extracted using Diethylene Triamine Penta Acetic acid (DTPA) following Lindsay and Norvell's (1978) method. The extract

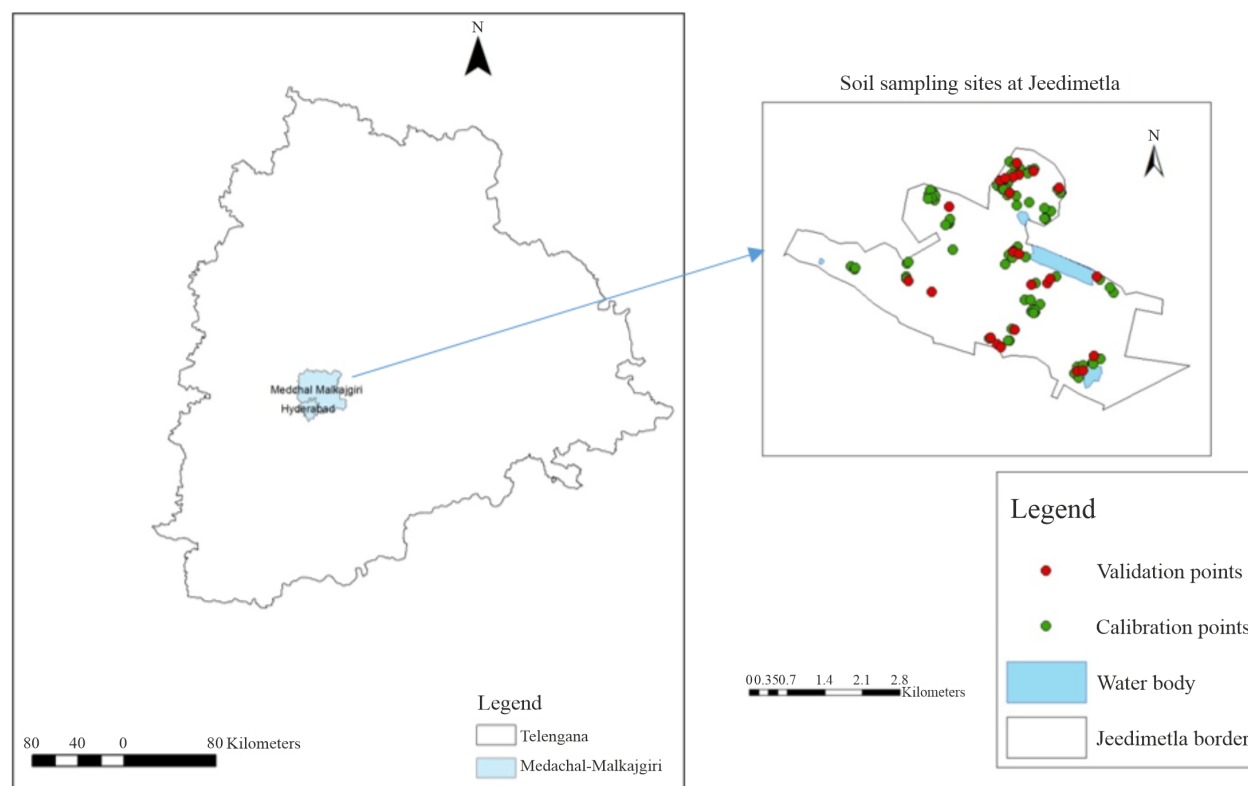


Fig. 1. Location of the study area and soil sampling sites

was fed to the instrument called ICP-OES (inductively coupled plasma-optical emission spectrometry) and measured the Mn concentration at 257.61 nm wavelength.

Spatial structure analysis and generation of spatial distribution map using geostatistics

The calibration dataset is used to generate spatial maps. This is done by using the simplest geostatistical tool called ordinary kriging. The dataset was initially used to compute a semivariogram model and then interpolated. The equation followed to compute the semivariogram is as follows:

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} \{Z(x_i) - Z(x_i+h)\}^2 \quad \dots(1)$$

where $\gamma(h)$ is semivariogram, $N(h)$ is the couple number of sampling points, $Z(x_i)$ is the observed value of the variable x at location i and $Z(x_i+h)$ is the observed value of variable x at distance h .

In the present study, spatial map was produced using Arc GIS 10.2 software by ordinary kriging method. Kriging is a sophisticated geostatistical tool that produces an estimated surface from a scattered set of points with z -values. Ordinary kriging assumes the model:

$$Z(s) = \mu + \varepsilon(s) \quad \dots(2)$$

(Where μ is the unknown, deterministic mean value, $\varepsilon(s)$ is a white noise process composed of measurement errors).

Accuracy assessment

The validation dataset is used for the accuracy estimation of the predicted map. It includes various validation indices such as coefficient of determination (R^2), root mean square error (RMSE), and mean absolute deviation (MAD). The details of the validation indices are as follows:

$$R^2 = \text{RSS} / \text{TSS} \quad \dots(3)$$

(Where RSS is sum of squares of residuals, TSS is total sum of squares.)

$$RMSE = \frac{\sqrt{\sum_{i=1}^N [y(i) - y'(i)]^2}}{N} \quad \dots(4)$$

(y is the predicted values and y' is the observed values, N is the number of validation points)

$$MAD = \frac{1}{n} \sum_{i=1}^n |x_i - m(X)| \quad \dots(5)$$

(Where $m(X)$ is average value of data set, n = number of data values, x_i is data values in the set)

Statistical analysis

The Pearson's correlation test between soil physicochemical properties and Mn content was carried out using IBM SPSS software. The significance level at $p < 0.05$ and $p < 0.01$ were considered.

Results and Discussion

Descriptive statistics of soil physico chemical properties

Red shallow soils were the predominant soil type of Jeeditmetla. The descriptive statistics of soil physicochemical characteristics, including pH, EC, and SOC, are presented in Table 1. The soil reaction analysis showed that the Jeeditmetla soils ranged from slightly acidic to moderately alkaline (5.63 and 8.9), with mean values of 7.28 and a standard deviation of 0.81. The agricultural land had the highest pH value of 8.9, while soil samples collected from the residential area had the lowest reading of 5.63. Electrical conductivity values ranged from 0.084 to 2.16 (dS m^{-1}), with a mean of 0.43 and a standard deviation of 0.3. The highest EC value (2.16 dS m^{-1}) was recorded in agricultural soil, whereas scrub/wasteland shows the lowest value. The oxidizable SOC content of the studied soil ranges from 0.14 to 2.16 (%). Agricultural soils were high in SOC content, whereas the soil samples collected from scrub/wasteland were low in SOC content.

Descriptive statistics of Mn concentrations, Normality test and critical limits

The soil samples were split into two sets 1) validation dataset, which included 25 samples, and 2) calibration dataset, which included 75 samples. The calibrated dataset showed that the Mn follows a normal distribution pattern (Table 2). The Mn

Table 1. Descriptive statistics of soil properties

Parameters	pH (n=100)	EC (dS m ⁻¹) (n=100)	Oxidizable organic carbon (%) (n=100)
Minimum	5.63	0.084	0.14
Maximum	8.9	2.16	2.16
Mean	7.28	0.43	0.92
SD	0.81	0.3	0.53
Skewness	-0.38	2.3	0.6
Kurtosis	2.24	14.58	2.63
Median	7.41	0.39	0.86
CV	0.11	0.70	0.58

Table 2. Descriptive statistics of Mn concentrations at Calibration site and Normality test

Parameters	Calibration site (n=75)	Validation site (n=25)
	Mn (mg kg ⁻¹)	Mn (mg kg ⁻¹)
Minimum	1.72	6.42
Maximum	64.2	66.7
Mean	26.6	28.5
SD	17.7	15.8
Skewness	0.77	0.43
Kurtosis	2.432	2.62
Median	23.05	27.08
CV	0.66	0.55
Distribution	Normal	NA

concentration in Jeeditmetla soil ranges from (1.72-64.2 mg kg⁻¹) with a mean value of 26.6 and a standard deviation of 17.7 mg kg⁻¹. The coefficient of variation was 0.66%. Kaur and Rani (2006) also reported higher available Mn concentrations (above

maximum permissible limit of 10 mg kg⁻¹) in Kanjhawala, western Najafgarh and Alipur soils of National Capital Territory (NCT), Delhi. Behera and Shukla (2013) also reported similar findings of DTPA-Mn in acid soils of Hariharapur, Debatoli, Rajpora and Neeleswaram in Orissa, Jharkhand, Himachal Pradesh and Kerala states of India, respectively. The land use-wise distribution of Mn is presented in Table 3. The Mn content was recorded highest in the soil collected from agricultural land with a value of 66.7 mg kg⁻¹. The mean value plus standard deviation of Mn content of soils of the residential area, industrial area, and scrub/wasteland were (22.2±13.9), (23.4±14.9), and (29.0±17.8), respectively. Overall there was a 49 to 64% variability observed in available Mn content in soils across the land uses. Similar observations were noted for validation datasets also with mean Mn content of 28.5 mg kg⁻¹ with SD and CV of 15.8 and 0.55, respectively.

Table 3. Descriptive statistics of available Mn concentrations in various land use

Parameters	Manganese (mg kg ⁻¹)			
	Residential area	Industrial	Agricultural land	Scrub/wasteland
Minimum	4.5	6.42	4.56	1.72
Maximum	63.3	60.2	66.7	64.2
Mean	22.2	23.4	38.3	29.0
SD	13.9	14.9	19.05	17.8
Skewness	1.23	1.09	-0.25	0.41
Kurtosis	4.27	3.55	2.01	2.01
Median	18.9	20.1	42.2	26.77
CV	0.62	0.64	0.49	0.61

Table 4. Correlation between soil properties and available Mn

	Mn (mg kg ⁻¹)	EC (dSm ⁻¹)	OC (g kg ⁻¹)	pH
Mn (mg kg ⁻¹)	1.00	0.20*	0.28**	-0.08
EC(dSm ⁻¹)	0.20*	1.00	0.25*	0.13
OC(g Kg ⁻¹)	0.28**	0.25*	1.00	-0.01
pH	-0.08	0.13	-0.01	1.00

** . Correlation is significant at the 0.01 level (2-tailed)

* . Correlation is significant at the 0.05 level (2-tailed)

Kaur and Rani (2006) and Singh *et al.* (2020) reported a <4, 4-10 and >10 ppm Mn content in soils as deficient, desirable and permissible limits. Results show that among 100 soil samples collected over the study sites only one sample falls under deficient, 18 samples under desirable and 81 samples beyond prescribed permissible limits of 10 mg kg⁻¹. The mean Mn concentration of the surface soils was 2 to 3 times higher than the prescribed permissible limits. However, on an average the soil samples collected from the agricultural lands shows 3 to 4 time's higher Mn concentration than the prescribed permishable limits.

Correlation between soil properties and DTPA extractable Mn

Correlation is a common method for determining the linear relationship between random variables. The association between different soil characteristics and Mn was examined in the current study using the Pearson correlation coefficient, with significance levels at (p<0.05) and (p<0.01). The results of the correlation test between Mn and soil characteristics such as SOC, pH, and EC are presented in table 4. Results show that Mn was strongly correlated with organic carbon and electrical conductivity at (p<0.01, r =0.28) and (p<0.01, r =0.20), respectively. However, Mn had a negative but not significant correlation with pH (r = -0.08).

Spatial structure of DTPA extractable Mn content

The degree of spatial continuity and spatial dependency can be assessed using semivariogram analysis. Selecting a suitable semivariogram is the

key to assess the spatial variability of a soil property. Semivariograms can be used to visualize the spatial variance of samples at different distances. The three parameters on which a semivariogram is described are nugget, sill, and range. In the present study, all the semivariogram models were explored, namely exponential, stable, circular, spherical, gaussian, etc., using ArcGIS geostatistical tool. The optimize model function was used to fit the semivariogram. Among all the models employed, the stable function showed (Fig. 2) better performance with a nugget to sill ratio of 0.51 (51%) and a range of 0.1 km. The result revealed that Mn showed a moderate spatial dependency. Tagore *et al.* (2015) reported a strong spatial dependency for Mn while predicting it using a spherical semivariogram model through ordinary kriging. However, Sharma *et al.* (2020) reported the exponential model was the best fit for Mn in black soils of central India. Ramzan and Wani (2018) have also reported exponential model as a best semivariogram fit for assessing spatial variability of available Mn content in soils. The strong and poor dependence is mainly caused by intrinsic and extrinsic factors (Camberdella *et al.*, 1994). The <25 and >75% values are considered a strong and poor spatial dependency, respectively (Mitran *et al.*, 2019). The variogram model parameters include nuggets representing the level of random variations within the database (Mitran *et al.*, 2018). Sill represents the magnitude of spatial variability within Mn datasets. In the present study, Mn showed a positive nugget value which indicates the combined effects of variability within the shortest sampling distance; hence measurement error is substantial. The ratio of nugget and sill showed the spatial dependency of Mn content in the studied soils.

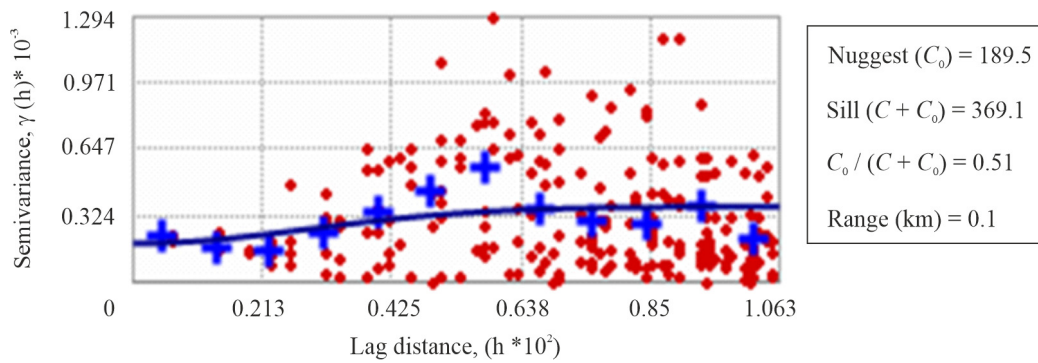


Fig. 2. Stable semivariogram model of Mn content

Spatial distribution Map of available Mn content

The ordinary kriging was employed to create a spatial distribution map over the study area using a stable semivariogram model presented in Fig. 3. The kriging predicted map observed a mean Mn content of 26.5 mg kg⁻¹ which is 2.5 times higher than the prescribed permissible limits in soils. The DTPA extractable Mn concentrations were observed to have a moderate spatial variable distribution pattern. The spatial distribution map of Mn showed that a higher concentration (>40 mg kg⁻¹) was observed in the study area’s northern and northeast parts. At the same time Mn concentration of <20 mg kg⁻¹ were recorded in the central and the western parts of the study area

mg kg⁻¹. However, Mn values of 25-40 mg kg⁻¹ were observed in the southeastern region of the study site. The spatial variability of available Mn concentrations in the study site could be largely attributed to the land use of the area.

Accuracy assessment

The accuracy assessment of the predicted Mn was carried out using validation sample points (25 sites). The validation indices are presented in Fig. 4. The accuracy was estimated based on the coefficient of determination (R²), mean absolute deviation (MAD) and root mean square error (RMSE). The accuracy assessment results show that the ordinary kriging with a stable variograms model could predict

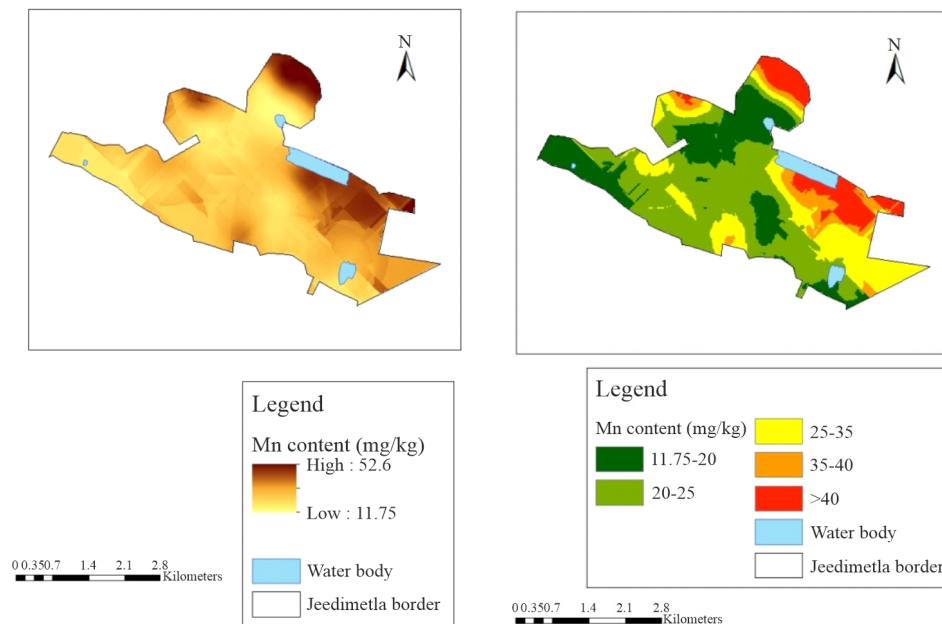


Fig. 3. Spatial distribution map of available Mn content of Jeedimetla soil

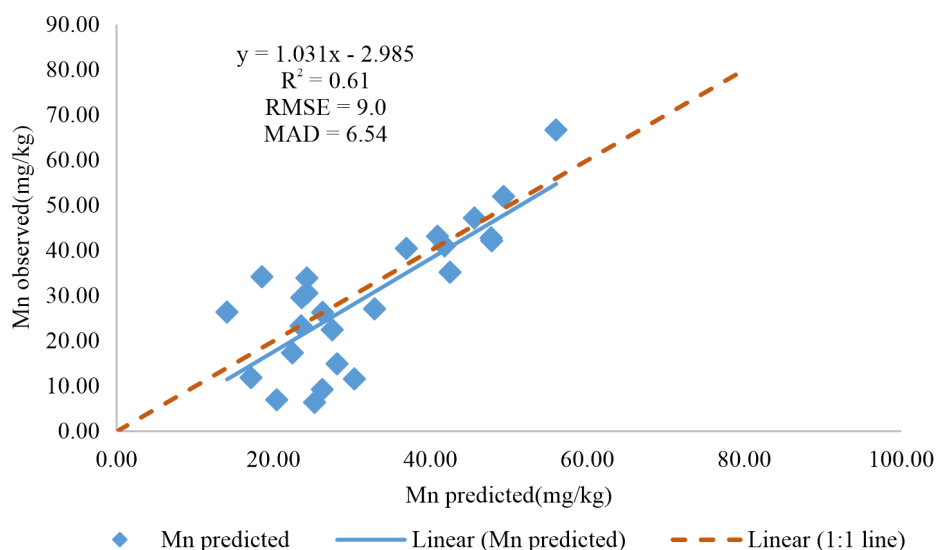


Fig. 4. Validation of ordinary kriging predicted soil Mn content map

Mn content in an unsampled location with moderate accuracy with R^2 of 0.61 and RMSE and MAD of 9.0 and 6.54 mg kg^{-1} , respectively.

Conclusions

In the present study, the geostatistical method, i.e., ordinary kriging was used to spatially predict DTPA extractable Mn content in soils of the Jeedimetla Industrial belt of Hyderabad. The stable semi-variograms function can capture moderate spatial dependency of Mn content in soils. Moderate prediction accuracy of 61% was achieved for Mn using ordinary kriging. However, there is a scope to explore other geostatistical, hybrid, machine learning approaches for spatial prediction of heavy metal content in soil. The kriging predicted map shows a mean Mn content of 26.5 mg kg^{-1} which is 2.5 times higher than the prescribed permissible limits in soils. Hence, the spatial Mn distribution map produced in the current study could be a valuable source of information on the spatial distribution pattern of Mn content in the study area, which can help in the identification of hotspots of Mn contamination in soils and the formulation of a suitable management plan which includes bioremediation, addition of organic matter, proper sewage and sludge treatment before disposal and balanced use of fertilizers etc. to maintain the concentration below critical limits.

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