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Quantification of trace elemental concentrations in ten soil samples from coal mining region by ED-XRF-technique

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ABSTRACT

In recent times, trace elemental analysis of various samples have an important role in human life. Ramagundam, one of the oldest coal mining areas in Telangana, India was explored for the soil trace elemental analysis. EDXRF technique was used to analyze the soil samples and quantification was also performed using the nEXT software. Nine soil samples were collected from coal mining area from Ramagundam and one soil sample was collected from agriculture area, which is considered as a control sample. Seven elements V, Cr, Mn, Fe, Ni, Cu, and Zn were found and the concentrations of Fe, Mn, Ni, V, and Cr were observed at the extreme level when compared with other elements. The elements V, Cr, and Ni are known to be toxic even at lower concentrations. These results were compared with NIST 2587 and control soil and found a wide variation in their elemental composition. The elements i.e., Zn, Fe, Cu, and Mn have a major role in human physiology, plant growth, and development. Since they are found to contain a substantial amount of micro-nutrients in these samples, it would lead to the betterment of the socio-economic life of the people living in the area of mining.

***Keywords*:** Elemental analysis, Trace elements, Soil samples from coal mine area, EDXRF spectrometer, nEXT-software

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1. INTRODUCTION

Coal is the most abundant fossil fuel on the earth and supplies about 75 % of the major fuel recourses [1]. On the other hand coal mining industry is one of the major concerns of environmental pollution throughout the world [2]. Coal mining activities are inevitably connected with production of acidic drainage and airborne compounds such as fly ash and bottom ash with high metal content [3]. The contamination of soils, water, and air with toxic metals may pose long term environmental and health implications. The adverse effect of coal mining on the environment as well as human health has been reported by many workers in different areas [2, 4] i.e., global warming by release of CH_4 gas, release of CO gas which pollutes air and mine workers health [5], and it also promotes soil erosion, dust pollution and respiratory problems in nearby colonies [6]. Fly ash and dust released from coal mining and burning activities which are causes to an increase of pulmonary and heart related deaths [7-8]. Coal sludge is caused for thermal water pollution when high temperature of this coal sludge mixed with nearby river water that shows adverse effect on river ecosystem and organisms also by reducing oxygen supply [9].

Top soil is an essential component in abandoned mines for the growth of vegetation and it has to be preserved for the post-mining land reclamation [10]. Thus understanding the occurrence, accessibility and ecological risk of trace elements present in around the coal mining area is of utmost important in view of implications for environmental health [11]. Trace elements in the soil are derived from the parent materials and various anthropogenic sources like urbanization and industrialization including mining and agriculture [12-13]. Among anthropogenic sources of trace elements, coal mining plays a significant role. Many toxic trace elements are released during coal mining and burning of coal [14], which are highly toxic to the human body even at trace amount.

The heavier elements like Cd, Pd, As, and Hg are highly toxic to the human body even at trace amount. Some of the trace elements can be transferred from soil to plant and may be adversely affect on ecology, crop growth, and agriculture production, enter the food chain and reach to the human body and ultimately harm to living organisms [15-17]. In this way, the coal mining industry has a negative impact on human health, environment, water and soil, causing their pollution and diseases. There are several methods that have been widely used for evaluating trace elemental concentration in soils. Nuclear analytical methods have a wide range of applications in chemical, biological, geological, toxicological, and environmental to characterize various types of materials. Several sophisticated techniques are utilized to determine major, minor and trace elements and their concentrations or elemental profile in different samples. Some of these techniques are XRF, AAS, NAA, ICP-MS, PIXE and EDXRF have been developed for trace elemental analysis [18-20]. Among these techniques Energy Dispersive X-Ray Fluorescence (EDXRF) is a special interest to be researchers as this technique is widely used for multi-elemental, non-destructive, sensitive, and relatively cheap while at the same time it enables simultaneously qualitatively and quantitative analysis of any size or number and without chemical pre-treatment [21]. In most case the emphasis has been given on the estimation of heavy metals in soils [22].

However, in the current study we focused only on the trace elemental composition in soil samples of Ramagundam mining area by EDXRF technique for the first time. Thus the research on soil contamination around coal mine sites should receive increasing attention in the restoration of soil ecosystems and their sustainable use. The purpose of soil analysis is to assess the adequacy, surplus, deficiency of various nutrients for vegetation growth and to monitor the changes that occurred during mining practices. The main aim of this study is to determine the trace elemental concentration present in ten selected soil samples from the coal mining area of Ramagundam.

2. EXPERIMENTAL METHODS

2. 1. Collection of soil samples

Ten soil samples for analysis were collected from Ramagundam coal mining area located in E 79°28'30'' and N18°45'50'', Peddapally district, Telangana State, India. For this two types of samples were collected, (a) nine samples from near to mining sites and (b) one is collected from agriculture area (control sample) shown in Figure 1. Each soil sample was collected from coal mine area, with a kilometer distance.

2. 2. Procedure of sample preparation

Figure 1. Simplified geological Google map of sampling locations of the study area of Ramagundam.

There are many methods are used for solid sample preparation including fusion, powder and etc., but in the present work, pellet samples to be analyzed by EDXRF-spectrometer. The fine powder samples prepared in the form of pellets have the advantage of high X-Ray intensity [22]. The samples were air dried, removed pebbles, extra moisture among them cleaned and subsequently grounded into a fine powder using an agate mortar and pestle. A quantity of 150 mille grams of each powder sample was taken to make the pellets with 13 mm diameter and 2 mm thickness by applying a pressure of 150 Kg/cm^2. Triplicates of each sample were prepared in a similar manner.

2. 3. Experimental set-up of EDXRF-analysis

The set-up consist of Xenemetrix (Previously Jordan Valley) EX-3600 spectrometer, Energy Dispersive X-Ray Fluorescence (EDXRF) spectrometer with Si (Li) detector resolution of 143 eV at 5.9 keV photo peak was used to obtain the elemental profile of concern soil from mine area. EDXRF spectrometer is designed to detect the elements in analyzed samples and determined their concentrations using X-Ray fluorescence. It is sensitive to elements from Z=11 (Na) to Z=99 (U). Samples are solid, liquid and powder or any other form and the detection limit of the set-up is 1-10 ppm. The empirical calibration technique was employed using Standard Reference Material NIST 2587. The in-built software nEXT was used for quantitative analysis [23]. To optimize the EDXRF sensitivities, different combinations of EDXRF parameters were employed. The different X-Ray filters were used to eliminate unnecessary radiation and noise [22]. For this present study, a 0.05 mm thickness Ti filter was used in front of the at a voltage of 14 kV was used for elements such as V, Cr, Mn, Fe, Ni, Cu and Zn measurements was done in 400-s [23].

The elemental analysis was performed using Mo K X-rays generated form a secondary molybdenum target of an EDXRF system available at Trace Elemental Laboratory, UGC, DAE-CSR Kolkata centre. The system incorporates a low power (50 W), air cooled Rh (Rhodium) anode X-Ray tube operated at 50 V and 1 mA power supply and 10 samples turret enables mounting and analyzing 10 samples at a time. These pellets were used as targets of EDXRF experiment. The spectra were recorded by using a PC based multi-channel analyzer. The spectral data were analyzed by computer program [24].

The elemental concentrations were determined using the calibration curve method by comparing the peak area and heights of the sample with that of certified reference material. Results were checked by Standard Reference Material (SRM) NIST 2587 for accuracy of the present investigation.

3. RESULTS AND DISCUSSION

The average elemental concentrations were determined in ten soil samples using EDXRF technique and thus the obtained results were shown in Table 1. Seven various elements such as Vanadium (V), Chromium (Cr), Manganese (Mn), Iron (Fe), Nickel (Ni), Copper (Cu) and Zinc (Zn) were found at different concentration levels. Figure 2 shows the bar graphs of elemental concentrations present in ten soil samples with certified reference material values of NIST 2587.

Analysis of present investigation shows a wide variation in their elemental concentrations in ten soil samples of mine area when compared with Certified Reference Material (CRM) values of NIST 2587. From the results it can be seen that the concentration of Fe, Mn, V, Cr and Ni were found at higher levels while the concentration of Cu and Zn were found at lower levels in selected different soil samples when compared with CRM vales of NIST 2587.

Table 1. Average trace elemental concentration (in ppm) of ten selected soil samples from coal mine area, Ramagundam, Telangana.

Soil samples	V	Cr	Mn	Fe	Ni	Cu	Zn
Sample 1	229.60	201.91	1219.26	76534.75	114.22	67.76	151.83
Sample 2	215.24	194.13	981.04	60364.25	93.17	61.53	149.61
Sample 3	187.66	141.21	958.11	44480.36	62.46	65.85	84.19
Sample 4	125.51	123.07	507.25	32147.82	40.65	29.13	81.93
Sample 5	114.36	100.95	440.76	28643.55	57.41	42.63	61.38
Sample 6	197.89	172.68	928.27	58068.98	80.18	54.18	62.30
Sample 7	127.67	148.08	666.63	56179.24	74.94	55.86	61.87
Sample 8	173.82	154.80	495.02	38261.66	70.24	61.53	72.44
Sample 9	114.26	190.63	147.78	46094.13	97.94	65.85	46.82
Control sample	79.77	91.32	458.42	29653.08	39.20	38.93	50.91
NIST 2587	78	92	651	28130	36	160	335.8

Here,

Vanadium (V)

In the present investigation the average concentration of Vanadium was varied from 215.24 ppm to 114.26 ppm. The concentration of V was found higher level in all ten soil samples and the appreciable amount of V was found in control soil sample when compared with CRM values of NIST 2587 i.e. 78 ppm. Vanadium occurs naturally in soil, water, and air. Natural sources of atmospheric V include continental dust, marine aerosol, and volcanic emissions. Releases of V to the environment are mainly associated with industrial sources, especially oil refineries and power plants using V rich fuel oil and coal. Global human-made atmospheric releases of vanadium have been estimated to be greater than vanadium releases due to natural sources. Natural releases to water and soil are far greater overall than human-made releases to the atmosphere [25]. No studies were located regarding the rate and extent of absorption in humans after oral exposure to vanadium. Adverse respiratory effects have been reported in humans and animals exposed to vanadium compounds at concentrations much higher than those typically found in the environment. Although the available data in humans are limited, signs of airway irritation (e.g., coughing, wheezing, sore throat) have been reported in subjects acutely exposed to 0.6 mg vanadium/m3 and in workers exposed to vanadium pentoxide dust.

Chromium (Cr)

In the present study the average concentration of Chromium was varied from 201.91 ppm to 100.95 ppm. The concentration of Cr was found very high in soil sample 1 i.e. 201.91 ppm and appreciable amount of Cr was found in control sample i.e. 91.32 ppm when compared with CRM vales of NIST 2587 i.e. 92.00 ppm. Some researchers have been reported that elemental analysis of soil samples from coal mining area situated in Hamadan province of Iron by PIXE technique [26]. Oswal *et al* found the average concentration of Cr to be70 ppm for nearby coal mine area and 47 ppm for control sample (clean area) [26]. Chromium is an essential element, required for carbohydrate and lipid metabolism, and its deficiency may be associated with cardiovascular disease. Long standing exposure with Cr acute dermatitis has been consistently reported in workers exposed Cr containing materials [27]. Excess Cr causes vomiting, central nervous system disorder, growth retardation and cancer in mans and animals [28].

Manganese (Mn)

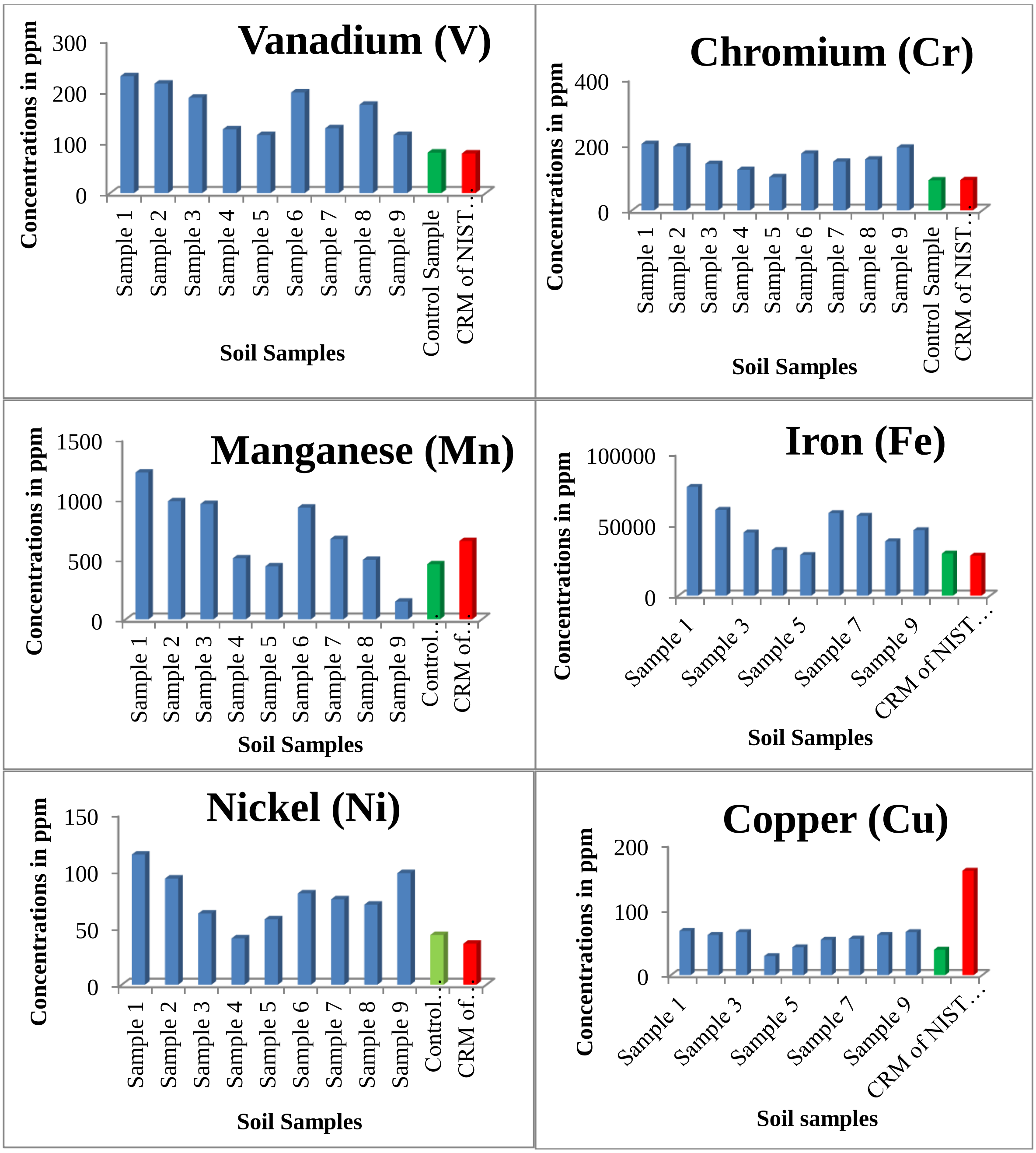
In the present study the average concentration of Manganese was varied from1219.26 ppm to 147.78 ppm. The concentration of Mn was found very high in soil sample 1 i.e. 1219.26 ppm while low concentration of Mn was found in soil sample 9 i.e.147.78 ppm when compared with CRM value of NIST 2587 i.e. 651 ppm. The appreciable amount of Mn was found in soil sample 7 i.e. 666.63 ppm. Oswal *et al* found that, the average concentration of Mn was 942 ppm for nearby coal mine and 193 ppm for control sample (clean area) [26]. Manganese acts as an activator of enzyme and as a component of metalloenzymes, play an important role in oxidative phosphorylation, fatty acids and cholesterol metabolism, mucopolysaccharide metabolism, and urea cycle [29].

Iron (Fe)

In the present study the average concentration of Iron was varied from 76534.75 ppm to 28643.55 ppm. The concentration of Fe was found higher in all ten soil samples except soil sample 5 when compared with CRM vales of NIST 2587. Fe has within the permissible limit in soil sample 5 and control soil sample. Oswal *et al* found that, the average concentration of Fe was 25294 ppm for nearby coal mine area and 11151 ppm for control sample (clean area) [26]. Fe is an essential element for human health especially for women. The excess amount of Iron cause rapid increase in pulse rate and coagulation of blood in blood vessels, hypertension and drowsiness and enormous excess of Fe may leads to metabolic and generic diseases [29].

Nickel (Ni)

In the present investigation the average concentration of Nickel was varied from 114.22 ppm to 39.20 ppm. The concentration of Ni was found higher amount in all ten soil samples and the appreciable amount of Ni was found in control sample i.e. 39. 20 ppm when compared to CRM values of NIST 2587 i.e. 36.00 ppm. The Ni was recorded above the maximum permissible limits of NIST 2587. Oswal *et al* found that, the average concentration of Ni was 48 ppm for nearby coal mine area and 20 ppm for control sample (clean area) [26]. Ni has essential trace element for human and animal health [30]. Ni is apparently involved in the protection of the cell membranes but actual requirements for human beings are not known. High level of Ni can be toxic and can leads to cancer [25].



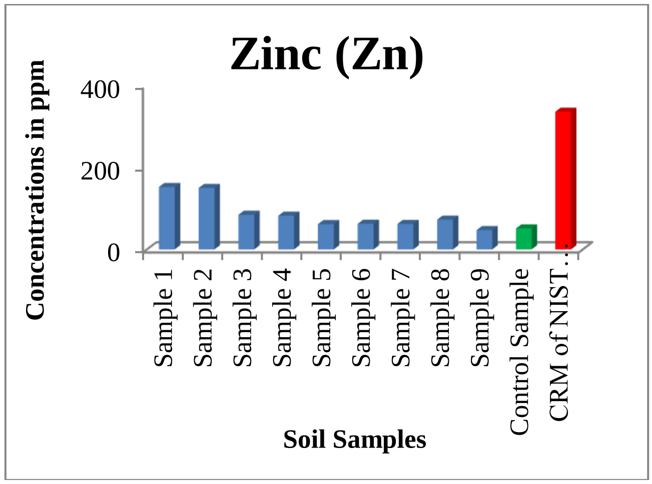


Figure 2. Bar-graphs of elemental concentrations of ten soil samples with CRM of NIST 2587

Copper (Cu)

In the present investigation the average concentration of Copper was varied from 67.76 ppm to 29.13 ppm. The concentration of Cu was found lower amount in all ten soil samples when compared with CRM values of NIST 2587 i.e. 160 ppm. Oswal *et al* found that, the average concentration of Cu was 31 ppm for nearby coal mine area and 12 ppm for control sample (clean area) [26]. Cu is essential for wide range of biochemical processes and it is necessary in diet as a trace element [27], it can be toxic at higher concentration. Cu toxicity is a fundamental cause of Wilson's disease [31].

Zinc (Zn)

In the present study the average concentration of Zinc was varied 151.83 ppm to 46.82 ppm. The concentration of Zn was found lower amount in all ten soil samples when compared with CRM vales of NIST 2587 i.e. 335.80 ppm. The control soil sample has also very low amount of Zinc i.e. 50.91 ppm. The concentration of Zn varied between 151.83 ppm to 46.82 ppm. Oswal *et al* found that, the average concentration of Zn was 53 ppm for nearby coal mine area and 21 ppm for control sample (clean area) [26]. Zn is one of the important trace elements and plays a vital role in physiological and metabolic processes of many organisms [30]. Zn deficiency is suspected to be common in developing countries. The deficiency of Zn negatively affects the healing of wounds, immune system response and ability to taste and smell.

From Table 2 it is clear that the average elemental concentration of Cr, Mn, Fe, Cu, Zn and Ni were found at higher levels in soil samples from nearby coal mine area whereas compared with both control sample as well as compared with the study of Oswal *et al* [26]. The variations were observed in the level of most of the trace elements in the soil samples from nearby coal mine and far away from coal mine. In both cases, the trace elemental concentrations were found at a higher amount in nearby coal mine areas, so this is due to different mineral compositions of soils from the mine area and these results indicate that the main sources of environmental pollution in Ramagundam are the emissions due to these heavy metal content.

Table 2. Comparison of present data with literature data.

Elements	Average elemental content of soils from Ramagundam by EDXRF (Present study)		Average elemental content of soils from Iron by PIXE [26]		Standard values of NIST 2587
	Soil samples nearby mine area	Control sample	Soil samples nearby mine area	Control sample	
Cr	158.60	91.32	70	47	92
Mn	704.90	458.42	942	193	651
Fe	48974.97	29653.08	25294	11151	28130
Cu	54.73	38.93	31	12	160
Zn	85.81	50.91	53	21	335.8
Ni	76.80	43.63	48	20	36

4. CONCLUSION

The trace elemental analysis was carried out in the control soil samples as well as selected soil samples from the coal mine area by EDXRF technique. Regular soil analysis for every 3-5 years is a vital part of good soil management practice [32]. From the present data, it is showed that the concentration of V, Cr, Mn, Fe, and Ni was found to be higher in all soil samples of coal mine area whereas Cu and Zn were found to be lower when compared with control soil sample and CRM values of NIST 2587. Most of the trace elements enumerating are relatively enormous excess amount which poses a more dangerous to people in within the area of mining.

The present study provides the essential information regarding the trace elements in soils, which elements enter the food chain and organic matter, and reach the human body than them harmful for public health. The present study also helps to reduce the magnitude of prevalence of essential trace element deficiency and toxicity diseases in the mining area.

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