Groundwater quality assessment with reference to some heavy metals toxicity and its probable remediation around Ballarpur area of Wardha valley coalfields, Maharashtra

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Abstract. Wardha valley coalfields are one of the major coal contributors in central India. The present study strives to assess the groundwater quality around Ballarpur area of Wardha Valley Coalfields concerning some heavy metals. 14 groundwater samples were taken for each pre-monsoon and post-monsoon season to assess Al, Cd, Cr, Cu, Fe, Pb, Ni, and Zn by AAS. Results disclosed that Cd, Fe, Pb and Ni were above the maximum permissible limit as per the Bureau of Indian Standards (2012). Fe, Zn, Cu and Al are the major percentage contributor in groundwater samples for both seasons. Shallow aquifer is more contaminated than deep. The PCA study has shown strong associations among a few heavy metals. The correlation coefficient study revealed that all heavy metals negatively correlate with pH values suggesting that contamination is favored in acidic water. Sampling locations and corresponding values imply mining activity as an influential cause for contamination. Hence, Acid Mine Drainage could cause a lower pH in the study area, augmenting heavy metals dissolution. To overcome, a flow system of anaerobic wetland and limestone drain could be a remedial method which will treat the mine discharge and mitigate the contamination.

1. Introduction

The quality of groundwater is among the most vital issues for human settlements and respective standards are established regarding water quality by various agencies depending upon locality and needs. The natural quality of groundwater is controlled by the lithosphere's geochemistry, the solid portion of the earth, and the hydrosphere's hydrochemistry, the aqueous portion of the earth [1]. In recent times the anthropogenic intervention has affected a lot to many groundwater regimes. Degradation and water pollution pollution are usually neglected until recent decades, but its suitability now has become a burning issue. Suitability of groundwater/surface water for a particular purpose depends upon the acceptable water quality standards for which it is being used [2].

The groundwater regime at many sites has suffered by intense coal mining, especially heavy metals. Several studies have shown that most of the contamination in coal mines can be released into the surrounding environment by leaching, and more attention should be paid to this kind of contamination [3]. Heavy metals are obstinate contaminants in groundwater as they cannot be degraded or removed and can halt for a substantially long period. Progressively they bio-accumulate by agricultural means and cause serious issues. Often they remain recessive, whereas, on disclosure to the positive situation,
they became active and are unrestricted to the outer environment. Although most major and trace metals are generally considered to be relatively immobile in the short term, their mobility under certain chemical conditions may exceed ordinary rates and pose a significant threat [4].

In the present study, groundwater quality in terms of heavy metals will be assessed in Ballarpur area of Wardha Valley coalfields. The study will reveal the contamination issue and the dissolution and migration process of heavy metals in the groundwater regime. The findings of the study may probably lead us to a practicable remediation policy over these contamination issues.

1.1. Study Area and Geology
The study area lies in Chandrapur district of Maharashtra with Latitude 19° 48’ 00” to 19° 56’ 00” N, and Longitude 79° 14’ 00” to 79° 23’ 00” E, traceable on toposheet no 56 M/6 (figure 1). The study area is experiencing continuous mining for many decades [5]. The topography of the study area is plain with alluvial deposits on both sides of the Wardha River. Sub-dendritic drainage is observed which signifies sedimentary terrain (figure 1.). The study area is a sub-basin of Godavari valley in Gondwana basin, located on the eastern limb of anticline plunging towards NNW [6]. Archean are the basement rocks majorly of quartzites, granite gneisses, etc. (table 1). In Gondwana rocks Kamthi Formation showing ferruginous red-brown sandstone and variegated shale are common, underlain by Barakar Formation where grey to white sandstone is common. Only one main coal seam also is known as Principal coal seam of the Wardha Valley is hosted by Barakar Formation which is high in ash content. Talchir formation with varieties of shale and sandstone is the lowermost in sequence and is overlain by Barakar Formation (table 1).

![Geology & Drainage Map](image)

**Figure 1.** Geology and drainage map of the study area with groundwater sample locations.
Table 1. General Geological succession from Wardha Valley Coalfield, Maharashtra, [7].

| Age                | Group/Formation          | Lithology                                              |
|--------------------|--------------------------|--------------------------------------------------------|
| Recent             | -                        | Alluvial gravel bed, black cotton soil                 |
| Eocene             | Deccan Trap              | Basalts                                                |
|                    | --------------- | ------      |
| Cretaceous         | Lameta Formation     | Limestone, charts and silicified sandstone              |
|                    | --------------- | ------      |
| Late Triassic      | Maleri Formation     | Fine to med. grained sandstone and red shale           |
| (To South East)    |                 |                                                        |
| Late Permian       | Kamthi Formation     | Red, brown sandstone, reddish siltstone and variegated shale & sandstone |
| Early Triassic     |                 |                                                        |
| Late Carboniferous | Talchir Formation    | Tillites, turbidites, varves, needles shale & sandstone |
| Early Permian      | Barakar Formation     | Light grey to white sandstone, shale & coal seam       |
|                    |                 |                                                        |
| Precambrian        | Sullavai Sandstone   | White and light brown quartzitic sandstone             |
|                    |                 |                                                        |
| Archean            | Pakhal Limestone     | Grey, bluish or pinkish limestone and chert            |

2. Methodology
In the study area, a collection of 14 representative groundwater samples for each pre-monsoon (May 2019) and post-monsoon (October-November 2019) were done (figure 1) in duplication. The sampling was done in 500 ml of narrow mouth pre-washed high-density polyethylene bottles. Before sampling, these bottles were rinsed with the sample water. The samples were acidified with 0.5 ml Concentrated HNO$_3$ to avoid any kind of chemical change through metal precipitation and were immediately transferred to an ice bath at approximately at 4°C, until analysis in the laboratory [8]. The bottles were air tightened and labeled in accordance with the locations. pH was measured on the spot by a digital meter to get an accurate reading. AAS was used to analyse Aluminium (Al), Cadmium (Cd), Chromium (Cr), Copper (Cu), Iron (Fe), Lead (Pb), Nickel (Ni), and Zinc (Zn). The obtained results were in µg/l. The percentage contributions of heavy metals were deduced by using the sum of average metal concentrations in the groundwater sample. Principal component analysis and correlation analysis were performed separately on the obtained data set by XLSTAT software to deduce various interrelationships. A formula was developed to identify the contamination intensity of contaminated groundwater samples.

3. Result and Discussion
The result obtained for heavy metals after analysis were correlated with the maximum permissible limit (in absence of alternate source) suggested by the Bureau of Indian Standards for drinking water [9]. The evaluation of the pre-monsoon and post-monsoon seasons data revealed many vital aspects of groundwater contamination. The principal component analysis, correlation analysis, and contamination intensity studies clarified the process of contamination to some extent.
3.1. Characterization of groundwater

The result obtained from pre and post-monsoon samples have revealed that 06 samples (GW3, GW4, GW8, GW9, GW10, and GW14) among 14 were contaminated concerning Cd, Fe, Ni, and Pb (table 2 & 3). The Cd concentration was found to be highest at GW10 for both seasons. The high Cd concentrations may be due to coal mining activity in the surrounding area. The maximum value for Fe was observed on GW3 for pre-monsoon and on GW8 for post-monsoon. The raised Fe concentration can be attributed to pyrite's oxidation present in rocks exposed due to mining. The concentrations of Ni and Pb were found to be highest on GW14 for both seasons. The observed high Ni concentration may be due to basaltic rocks and shale, whereas the high Pb concentration could be due to shale and igneous rocks [1].

Table 2: The results obtained for heavy metals from pre-monsoon groundwater samples.

| Sample | Locations | pH  | Al  | Cd  | Cr  | Cu  | Fe  | Ni  | Pb  | Zn  |
|--------|-----------|-----|-----|-----|-----|-----|-----|-----|-----|-----|
|        |           | ρ  | * (200) | (3) | (50) | (1500) | (300) | (20) | (10) | (15000) |
| GW1    | 19°54'07"N 79°14'47"E | 6.2 | 45 | ∞ | 40 | 45 | 110 | ∞ | 5 | 30 |
| GW2    | 19°53'15"N 79°14'14"E | 6  | 90 | ∞ | 38 | 140 | 225 | 15 | 10 | 220 |
| GW3    | 19°55'44"N 79°18'14"E | 5.2 | 165 | 12 | 45 | 110 | 460 | 45 | 40 | 515 |
| GW4    | 19°54'02"N 79°18'24"E | 5.4 | 110 | 32 | 42 | 75 | 320 | 32 | 25 | 220 |
| GW5    | 19°55'33"N 79°19'18"E | 6  | 75 | ∞ | 25 | 47 | 188 | ∞ | 7 | 40 |
| GW6    | 19°51'59"N 79°20'10"E | 5.9 | 66 | ∞ | 20 | 66 | 300 | 10 | ∞ | 33 |
| GW7    | 19°53'03"N 79°20'29"E | 5.9 | 85 | ∞ | 35 | 60 | 170 | ∞ | 9 | 20 |
| GW8    | 19°50'48"N 79°21'29"E | 5.1 | 140 | 20 | 42 | 220 | 430 | 50 | 30 | 270 |
| GW9    | 19°49'48"N 79°19'51"E | 4.9 | 120 | 18 | 40 | 130 | 382 | 38 | 45 | 120 |
| GW10   | 19°49'10"N 79°18'24"E | 5  | 85 | 40 | 52 | 105 | 440 | 55 | 39 | 400 |
| GW11   | 19°51'31"N 79°17'28"E | 5.9 | 49 | ∞ | 20 | 70 | 70 | ∞ | 5 | 20 |
| GW12   | 19°51'03"N 79°16'04"E | 6  | 75 | ∞ | 18 | 63 | 130 | ∞ | 10 | 39 |
| GW13   | 19°49'40"N 79°14'56"E | 6.2 | 120 | ∞ | 14 | 40 | 115 | 9 | ∞ | 68 |
| GW14   | 19°49'27"N 79°16'27"E | 5.2 | 102 | 25 | 48 | 320 | 280 | 60 | 70 | 395 |
| Min.   |           | 4.9 | 45 | ∞ | 14 | 40 | 70 | ∞ | ∞ | 20 |
| Max.   |           | 6.2 | 165 | 40 | 52 | 320 | 460 | 60 | 70 | 515 |
| Arithmetic Mean |     | 5.63 | 94.78 | 10.50 | 34.21 | 106.50 | 258.57 | 22.42 | 21.071 | 170.71 |
Table 3: The results obtained for heavy metals from post-monsoon groundwater samples.

| Sample | Locations | pH | Al | Cd | Cr | Cu | Fe | Ni | Pb | Zn |
|--------|-----------|----|----|----|----|----|----|----|----|----|
| GW1    | 19°54'07"N 79°14'47"E | 6.6 | 30 | ∞  | 32 | 30 | 105 | 7  | 3  | 21 |
| GW2    | 19°53'15"N 79°14'14"E | 6.5 | 100 | ∞ | 24 | 80 | 225 | 10 | 8  | 98 |
| GW3    | 19°55'44"N 79°18'14"E | 6  | 110 | 7  | 33 | 120 | 330 | 33 | 16 | 325|
| GW4    | 19°54'02"N 79°18'24"E | 5.9 | 124 | 20 | 50 | 72 | 304 | 40 | 20 | 225|
| GW5    | 19°55'33"N 79°19'18"E | 6.2 | 55  | ∞ | 21 | 45 | 155 | ∞  | 6  | 55 |
| GW6    | 19°53'59"N 79°20'10"E | 6  | 40  | ∞ | 15 | 68 | 280 | 5  | 5  | 24 |
| GW7    | 19°53'03"N 79°20'29"E | 6.4 | 90  | ∞ | 30 | 71 | 144 | 8  | ∞  | 31 |
| GW8    | 19°50'48"N 79°21'29"E | 5.6 | 130 | 12 | 45 | 163 | 368 | 36 | 21 | 241|
| GW9    | 19°49'48"N 79°19'53"E | 5  | 80  | 10 | 22 | 108 | 322 | 42 | 33 | 106|
| GW10   | 19°49'10"N 79°18'24"E | 4.8 | 72  | 24 | 50 | 110 | 300 | 40 | 14 | 387|
| GW11   | 19°51'31"N 79°17'28"E | 6.4 | 30  | ∞ | 15 | 58 | 50  | ∞  | ∞  | 12 |
| GW12   | 19°51'03"N 79°16'04"E | 6.3 | 60  | ∞ | 22 | 50 | 47  | ∞  | 8  | 41 |
| GW13   | 19°49'40"N 79°14'56"E | 6  | 105 | ∞ | 18 | 40 | 125 | 10 | ∞  | 60 |
| GW14   | 19°49'27"N 79°16'27"E | 5.7 | 143 | 10 | 35 | 224 | 365 | 57 | 74 | 262|
| Min.   |           | 4.8 | 30  | ND | 15 | 30 | 47  | ∞  | ∞  | 12 |
| Max.   |           | 6.6 | 143 | 24 | 50 | 224 | 368 | 57 | 74 | 387|
| Arithmetic Mean | 5.90 | 83.50 | 5.92 | 29.42 | 88.50 | 222.85 | 20.57 | 14.85 | 134.85 |

ρ - Maximum permissible limit by BIS (2012). ∞ – Not Determined

3.2. Interpretations

3.2.1. Percentage contribution. The contribution of each heavy metal in groundwater samples for pre-monsoon was calculated based on their average in pre-monsoon. The observed percentage contributions are Al (13 %), Cd (02 %), Cr (05 %), Cu (15 %), Fe (36 %), Ni (03 %), Pb (03 %), and Zn (23 %) (figure 2a). In post-monsoon samples the percentage contribution of each metal is observed as Al (14 %), Cd (01 %), Cr (05 %), Cu (15 %), Fe (37 %), Ni (03 %), Pb (02 %), and Zn (23 %) (figure 2b). The Al and Fe contribution has increased by 1%, and the contribution of Cd and Pb has decreased by 1% in post-monsoon. The increased percentage of Al and Fe may be due to increased dissolution form surficial anthropogenic sources, whereas the decreased value of Cd and Pb could be the dilution effect.
3.2.2. **Principal Component Analysis.** The principal component analysis has revealed that the 82.74% variance in pre-monsoon and 85.65% variance in post-monsoon are attributed to the first two factors. These factors are then plotted to deduce the similarities and dissimilarities among variables and observations. The biplot revealed positive relationships among heavy metals in both seasons. In pre-monsoon, the vectors indicate Cd, Cr and Pb show a strong positive correlation, whereas Cu–Ni–Zn–Cd with reasonable positive correlation (figure 3a). The Cu–Ni–Zn–Cd association can indicate the origin from igneous and sedimentary rock types [1]. The weakest positive correlation can be found between Al and Cu. In post-monsoon, the vectors for metals are more dispersed, indicating comparatively weak relationships when compared to pre-monsoon. The vectors of Ni-Fe and Cr-Cd are closely related, whereas Cr-Pb is least correlated in the post-monsoon (figure 3b). The Samples are grouped into two halves where one is contaminated, and the rest are non-contaminated for both seasons.

**Figure 2.** Percentage contribution of each heavy metal in groundwater sample for a. pre-monsoon and b. post-monsoon.
3.2.3. Contamination Intensity in groundwater samples. The results have clarified that the groundwater regime is contaminated by Cd, Fe, Ni, and Pb. To understand the severity and distribution of these heavy metals in contaminated samples, the contamination intensity was deduced by the following formula for every metal having concentration above the maximum permissible limit as per BIS (2012).

**Figure 3.** Factor biplots of observations and variables for a. pre-monsoon and b. post-monsoon.
The values obtained in the form of $\Upsilon$ for Cd, Fe, Ni and Pb (table 4), provided the least and highest contaminated location in the study area. This data can be used to sort the contaminations source location and plan for further remedial actions. Based on the average $\Upsilon$ value (table 4), GW10 was the most contaminated sample and GW3 with the least contamination in pre-monsoon samples (figure 4a). Whereas in post-monsoon, GW14 was highest in contamination level, and again GW3 with the lowest contamination (figure 4b). Shallow water aquifer is more contaminated than deep as GW8, GW10, and GW14 are shallow water samples.

**Table 4.** Contamination intensity ($\Upsilon$ values) for heavy metals found above the maximum permissible limit as per BIS (2012) in groundwater samples for both seasons.

|       | GW3  | GW4  | GW8  | GW9  | GW10 | GW14 |
|-------|------|------|------|------|------|------|
| Cd    | 4    | 2.33 | 10.6 | 6    | 6.66 | 8    |
|       |      |      |      |      | 3.33 | 8    |
|       |      |      |      |      |      | 8.3  |
|       |      |      |      |      |      | 3.33 |
| Fe    | 1.53 | 1.1  | 1.06 | 1.01 | 1.43 | 1.22 |
|       |      |      |      |      | 1.27 | 1.07 |
|       |      |      |      |      | 1.46 | 1.01 |
|       |      |      |      |      | 1   | 0.93 |
|       |      |      |      |      | 1.21 |
| Ni    | 2.25 | 1.65 | 1.6  | 2    | 2.5  | 1.8  |
|       |      |      |      |      | 1.9  | 2.1  |
|       |      |      |      |      | 2.75 | 2    |
|       |      |      |      |      | 3    | 3.85 |
| Pb    | 4    | 1.6  | 2.5  | 2    | 3    | 2.1  |
|       |      |      |      |      | 4.5  | 3.3  |
|       |      |      |      |      | 3.9  | 1.4  |
|       |      |      |      |      | 7    | 7.4  |
| Avg.  | 2.945| 1.67 | 3.95 | 2.9175 | 3.397 | 2.28 |
|       |      |      |      | 3.4175 | 2.45 | 5.36 |
|       |      |      |      | 5     | 3.1  | 4.8075 |
|       |      |      |      | 3.6975 | 5    | 5    |

[$\Upsilon = \frac{\text{Observed Metal Concentration}}{\text{Maximum Permissible Limit}}$]

$\Upsilon$ – Contamination intensity in groundwater sample.
3.2.4. Correlation coefficient studies. Coalfield’s drainage somehow controls the pH of the study area. Acid mine drainage is a widespread issue in coalfields. Hence the relationship between pH and contamination cannot be neglected, especially in coalfields. The correlation coefficient analysis for pre-monsoon has revealed the negative relationship between the samples pH value and their corresponding metal concentrations (figure 5). This indicates that the lower pH enhances the metal solubility and stability in groundwater. The correlation coefficients among heavy metals were reasonably high, indicating strong positive relationships (table 5).

Table 5. Correlation coefficient matrix for pre-monsoon groundwater parameters

| Parameters | pH  | Al  | Cd  | Cr  | Cu  | Fe  | Ni  | Pb  | Zn  |
|------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| pH         | 1   |     |     |     |     |     |     |     |     |
| Al         | -0.60 | 1   |     |     |     |     |     |     |     |
| Cd         | -0.84 | 0.39 | 1   |     |     |     |     |     |     |
| Cr         | -0.73 | 0.37 | 0.74 | 1   |     |     |     |     |     |
| Cu         | -0.61 | 0.39 | 0.48 | 0.56 | 1   |     |     |     |     |
| Fe         | -0.87 | 0.68 | 0.72 | 0.69 | 0.45 | 1   |     |     |     |
| Ni         | -0.91 | 0.64 | 0.86 | 0.77 | 0.75 | 0.84 | 1   |     |     |
| Pb         | -0.86 | 0.52 | 0.75 | 0.75 | 0.80 | 0.66 | 0.89 | 1   |     |
| Zn         | -0.72 | 0.66 | 0.70 | 0.75 | 0.61 | 0.77 | 0.88 | 0.77 | 1   |
Figure 5. Plots for correlation coefficient studies of pH in groundwater with: a. Aluminium, b.
Cadmium, c. Chromium, d. Copper, e. Iron, f. Nickel, g. Lead and h. Zinc for pre-monsoon.
In post-monsoon, the pH values and corresponding metal values are in a negative relationship, but with milder strength compared to pre-monsoon relationships (figure 6). All metals are in milder positive relationships, with very few of them indicating strong associations (table 6).

**Table 6.** Correlation coefficient matrix for post-monsoon groundwater parameters.

| Parameters | pH   | Al   | Cd   | Cr   | Cu   | Fe   | Ni   | Pb   | Zn   |
|------------|------|------|------|------|------|------|------|------|------|
| pH         | 1    |      |      |      |      |      |      |      |      |
| Al         | -0.30| 1    |      |      |      |      |      |      |      |
| Cd         | -0.78| 0.46 | 1    |      |      |      |      |      |      |
| Cr         | -0.44| 0.53 | 0.85 | 1    |      |      |      |      |      |
| Cu         | -0.51| 0.69 | 0.50 | 0.45 | 1    |      |      |      |      |
| Fe         | -0.66| 0.65 | 0.67 | 0.56 | 0.78 | 1    |      |      |      |
| Ni         | -0.73| 0.71 | 0.81 | 0.68 | 0.82 | 0.85 | 1    |      |      |
| Pb         | -0.47| 0.60 | 0.44 | 0.33 | 0.87 | 0.66 | 0.82 | 1    |      |
| Zn         | -0.66| 0.61 | 0.84 | 0.78 | 0.68 | 0.75 | 0.81 | 0.51 | 1    |

![Graphs](image)
3.2.5. Ingestion of heavy metals into groundwater.

The high Fe concentration indicates pyrite's oxidation within coal measures and associated strata and removal of oxidation product by inflowing groundwater [10]. Sulphuric acid (H2SO4) is formed when leached sulphur from pyrite oxidation reacts with water and atmosphere, which subsequently produces acid mine drainage (AMD). This AMD ultimately lowers the pH of the Groundwater [11]. On rock-water interaction, heavy metals present in the rock strata get dissolved in the groundwater and migrate to the local water regime. These heavy metals exist in associations like sulfides of lead and cadmium, which would naturally be found occurring together with sulfides of iron (pyrite, FeS2) and copper (chalcopyrite, CuFeS2) as minors [12].

3.3. Probable remedial measures

The groundwater regimes around coalfields are under the abundant risk of contaminations due to continuous mining activities. The interaction of water with exposed rocks in mines alters the chemistry of water, affecting groundwater drastically. The drainage from coal mine environments is usually acidic, also known as acid mine drainage. The acid mine drainage is an environmental pollutant that impairs water resources in the mining region throughout the world [11]. The contamination issue can be achieved by acidity check of the mine drainage before releasing it into a nearby water system [13]. A flow system of wetland and limestone beds could be a mechanism to treat AMD. This method is cheap and eco-friendly, with low maintenance costs. The mine water has to be released in the flow system to check the acidity of mine drainage. An artificially prepared anaerobic wetland will drive a sulfate reduction process to form a solid-phase metal sulfide as an alternative end product. This solid-phase sulfide will
remove metals from the solution and deposit them in the substrate (eq. 1) [12]. The subsequent bicarbonate (HCO₃⁻) will enhance the alkalinity by reducing H⁺ ions (eq. 2).

\[
M \text{(Metal)} + SO_4^{2-} + CH_3O \rightarrow MS + HCO_3^- \tag{1}
\]

\[
\text{HCO}_3^- + H^+ \rightarrow H_2O + CO_2 \text{(aq)} \tag{2}
\]

The limestone bed underneath the wetland will increase alkalinity by CaCO₃ dissolution (eq. 3). Finally, the treated drain will be safe to release into the nearby water bodies.

\[
\text{CaCO}_3 + H^+ \rightarrow Ca^{2+} + HCO_3^- \tag{3}
\]

4. Conclusion
The groundwater samples assessment of both seasons has revealed that 06 among 14 samples were above the maximum permissible limit as per BIS (2012) with respect to Cd, Fe, Ni, and Pb. The pre-monsoon samples showed higher contaminations than post-monsoon samples. This signifies that the water paucity in the system favors contamination. Fe, Zn, Cu, and Al are in major concentration in groundwater samples of both seasons. Shallow groundwater samples were found to be more contaminated than deep. The sampling locations and corresponding metal values indicate that the mining activity is responsible for the enhanced metal concentrations. The PCA studies grouped the samples and heavy metals as per their associations, which will ultimately provide an advantage for source finding studies. The correlation studies revealed the strong negative relationships of sample pH with respective metals concentrations. Thus the acidity favours the metal dissolvability is confirmed. All the above observations point to acid mine drainage's pivotal role in the dissolution and migration of heavy metals. High Fe concentration concludes oxidation of Pyrite (FeS₂) simultaneously, resulting in acidic drainage. The remediation of the contamination issue, a flow system with an anaerobic wetland followed by a limestone bed, is suggested. This suggested system will improve the alkalinity and hold the dissolved metals in the solid phase by a natural reducing environment.

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