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Trace elements contamination in groundwater and associated human health risk in the industrial region of southern Sonbhadra, Uttar Pradesh, India

Arif Ahamad · N. Janardhana Raju · Sughosh Madhav · A. H. Khan

Abstract The present study assesses the pollution load of the groundwater with reference to the trace elements (i.e. As, Hg, Cd, Cr, Cu, Fe, Mn, Zn, Ni, Co and Pb) and the potential health risk by its consumption for the residents of Obra, Renukoot and Anpara industrial clusters of Southern Sonbhadra, Uttar Pradesh, India. For this, 220 groundwater samples were collected during post- and premonsoon seasons in 2015. pH varied from slightly acidic to alkaline in both the seasons. Geochemical analysis of the area showed that all the three clusters are severely contaminated with Fe, Pb, Cd, Cr, As and Hg during both the seasons. High concentration of heavy metals indicates that groundwater was contaminated with natural as well as anthropogenic sources. For all the three clusters, the mean values of heavy metal pollution index were found above the critical index in both the seasons with Anpara in lead. For the majority of groundwater samples across the clusters during both the seasons, substantial non-cancer health risk was observed due to target hazard quotient values of Cr, Cd, As, Pb and Hg higher than unity. The hazard index value for children was very high compared to adults which means that children are more susceptible to health impairment in terms of non-carcinogenic health risk. Carcinogenic risk was higher for adults than children in the entire study area.

Keywords Heavy metals · Groundwater · Heavy metal pollution index · Target hazard quotient · Sonbhadra · Cancer risk

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Introduction

Today heavy metal pollution has invited greater attention because of its harmful nature and impact on the human and aquatic health especially as they continue exceed the prescribed standard permissible limits (Adamu et al. 2014; Jiménez-Ballesta et al. 2017). UNEP (2000) has listed some of the major anthropogenic sources of toxic heavy metal pollution in the aquatic environment including mineral extraction, sewage disposal, agrochemicals (pesticides) used in agriculture and discharges of untreated and semi-treated effluents from metal-related industries (smelters). Many researchers have revealed that the thermal power plants, by their combustion and fusion of coal, release different toxic constituents into the

environment such as ash pond leachate and fly ash (Usmani and Kumar 2017; Sarode et al. 2010; Gao et al. 2013; Nanos et al. 2015). The greatest victims of these plants are the water bodies present nearby since they are the ends of industrial effluents in the developing countries. The pollution load of the heavy metals is carried in the water run-off from the coal washers which contaminates the groundwater resources, lakes and rivers and further affects the aquatic flora and fauna (Finkelman 2007). The coal ash is a mineral residue after the burning of coal. It contains many toxic elements including arsenic, mercury, cadmium, lead, selenium, chromium and nickel (Lemly 2018). Indiscriminate discharge of effluent from chlor-alkali plant and cement industries enhances the mercury contamination (Sikkema et al. 2011; Sahu et al. 2014). In Northern Mongolia, some studies have attributed mining activities apart from the burning of coal having arsenic traces for the increase in level of arsenic in the surface and groundwater (Hofmann et al. 2010; Kasimov et al. 2011; Inam et al. 2011; Murao et al. 2011, Thorslund et al. 2012; Batbayar 2012; Pfeiffer et al. 2015). Pb and Cd contamination has been reported in the groundwater in and around Yatagan thermal power plant in Turkey (Baba et al. 2003). The presence of higher concentration of heavy metals like Fe, Ni, Al, Mn, Pb and As has also been observed in water bodies near Chhattisgarh's Korba coalfield (Singh et al. 2017) and in Tamil Nadu's lignite mines (Khan et al. 2005). The groundwater quality deterioration is caused by several processes such as mineral weathering, geochemical reactions, dissolution/precipitation reaction and ion exchange (Raju et al. 2009, 2016; Patel et al. 2018; Ahamad et al. 2018a; Madhav et al. 2018). Amongst all the pollution indices and after the assessment of toxic trace elements, heavy metal pollution index (HPI) has been found to be the most effective technique in rating water quality (Edet and Offiong 2002; Bhuiyan et al. 2010). Several studies, in India and as well as across the world, have been carried out to assess the contamination caused by toxic trace elements using pollution indices (Mohan et al. 1996; Prasad and Jaiprakas 1999; Prasad and Bose 2001; Giri et al. 2010; Kumar et al. 2012; Yankey et al. 2013; Giri and Singh 2014; Guan et al. 2014; Jahanshahi and Zare 2015; Panigrahy et al. 2015; Tiwari et al. 2015, 2017). Several researchers used non-carcinogenic and carcinogenic risk indices to evaluate the possible human

health risk due to the exposure to heavy metals in the different regions of the world (Wu et al. 2009; Storelli et al. 2010; Khan et al. 2013b; Gupta et al. 2015; Pawar and Pawar 2016; Singh et al. 2018; Ahamad et al. 2018b).

The southern part of Uttar Pradesh's Sonbhadra District and the north-eastern part of Madhya Pradesh's Singrauli District are popularly known as the "Energy capital of India". It is called so because of the presence of power grade coal reserves along with availability of water resources from Govind Ballabh Pant Sagar (G.B.P.S.) water reservoir (Rihand Dam). This provides an excellent location for other coal dependent industries like thermal power stations, aluminium smelting, cement industries, chlor-alkali industry, hi-tech carbon industry and other industrial and commercial operations adjacent to the coal mines (Khan et al. Khan. 2013a; Das et al. 2018). The surface and groundwater of Singrauli are severely contaminated due to direct pouring of fly ash slurry and leaching of toxic trace elements like As, Se, Hg, Cd and Pb from ash disposal pond of the power plant, making the water resources unfit for uses (Pandey et al. 2011). In Sonbhadra region, few studies have been done on heavy metals (Cd, Pb, Ni, As, Hg) in soil, sediment, aquatic biota and drinking water in and around Anpara coal-fired power plant (Agrawal et al. 2010, 2011) and Govind Ballabh Pant Sagar Reservoir (Rai 2010; Sahu et al. 2014). A study of environmental NGO, centre for science and environment found symptoms related to exposure of mercury in the local population. They reported that these symptoms appeared in the population due to consumption of contaminated water and food in the study area. An article published earlier in 2015 in 'Down To Earth' magazine brought forward this issue of mercury contamination. It leads us to conduct this research work in the study area. The populations living around these industrial clusters use groundwater as their primary drinking source. The groundwater laden with these toxic heavy metals may pose serious health issues. No study has been done on human health risk assessment due to exposure of heavy metal via consumption of groundwater in the area. Hence, the following objectives were set to carry out this research (a) distribution of heavy metals in groundwater and their seasonal variation around the all three industrial clusters, (b) determination of pollution load with the help of heavy metal pollution index (HPI) around all

the three industrial clusters and (c) investigation of non-carcinogenic and carcinogenic human health risks on both adults and children due to exposure of trace metals through groundwater ingestion pathways.

Study area

Geographically, the district is located between 23°52′ and 24°55′ N latitude and 82°30′ to 83°33′ E longitude. For the present work, three industrial clusters comprising of Obra (24°26′30.3″–24°28′41.10″N and 82°57′41.20″–83°02′48.90″E), Renukoot (24°11′17.20″–24°14′21.72″N and 83°00′22.50″–83°03′39.10″E) and Anpara (24°07′23.76″–24°11′30.30″N and 82°41′48.12″–82°47′36.30″E) having 55.50, 45 and 89.40 km^2 area, respectively, were selected. The study area is a hub for various industrial and commercial operations. The different industries present in the area are summarized as follows: Obra (Cluster A: Thermal Power Plant, Cement Factory, Sandstone–limestone Mining), Renukoot (Cluster B: Chlor-alkali Plant, Aluminium Smelting Plant and Hi-tech Carbon Limited) and Anpara (cluster C: Major Coal Mining and Thermal Power Plant). The total population of Sonbhadra District, as per the 2011 census is 1,862,559. The three industrial clusters of the study comprise of 113,815 population in which 15,809 are children and remaining are adults. Their location can help to understand the industrial impact on the groundwater resources (Fig. 1) of the area which has a typical dry-tropical climate and where the average rainfall fluctuates between 850 and 1300 mm. Mean monthly minimum temperature of the area varies between 13.4 and 30.7 °C, whereas the mean monthly maximum ranges between 23.4 and 40.2 °C. The general topography of the area is undulating, interspersed with flats, hills and valleys. The hilly tract in the southern part of the study area is almost parallel to the Ganga-Yamuna lineament. Geology of the study area comprises diverse rock formations ranging from archaean (Bundelkhand Granitic gneisses) to recent (Ganga) alluvium. The tract lies underneath Bundelkhand region's granitic complex and Sonbhadra's overlaying the rocks of mahakoshal (bijawar) and vindhyan supergroup. In the south-western Sonbhadra, while the younger rocks are constituted of the coal bearing gondwana, the lower vindhyan sediments of Sonbhadra are composed from

deposits of the cement grade limestone, flux grade dolomite and building stone.

Singrauli, a part of study region, was ranked ninth amongst most critically polluted areas of India with an index of 81.8 out of 100 (CPCB 2009). The study area was highly under developed and unexploited until the construction of the Rihand Dam (Govind Ballabh Pant Sagar) in 1961, over the river Rihand, a tributary of the Son River. Since then coal mining activities have increased extensively with most of the coal mines located near Uttar Pradesh and Madhya Pradesh border. Currently, around 83 million tonnes of coal is mined from fourteen mines per year. Alongside, many coal-fired thermal power plants have been established which contribute to about 12,700 MW thermal power (from ten power plants) in the area.

Materials and methods

Sample collection and processing

A total of 220 groundwater samples in 125 ml Tarson polyethylene bottle were collected from borewells and hand pumps during postmonsoon (January 2015) and premonsoon seasons (June 2015) from the three clusters in the study area, i.e. Obra (92 samples: 46 each season), Renukoot (72 samples: 36 each season) and Anpara (56 samples: 28 each season) for pH, electrical conductivity (EC) and trace elements analysis. Groundwater sampling was done before and after the monsoon to check variation amongst different parameters (whether their concentration are increasing or decreasing) and the pollution load and health risk so that the remedial measures can be planned accordingly by the decision makers. The groundwater samples were randomly collected from the functional borewells apart from the hand pumps around the industrial site that have been primarily used for drinking and domestic purposes by the residents. The study used the standard methods of APHA (1995) for the collection, preservation and analysis of the groundwater samples. For locating each sampling site, global positioning system (GPS) was used. For the metal analysis, the samples of groundwater were collected and preserved in the polyethylene containers by adding ultrapure HNO_3 except for mercury, the samples for which were collected in glass bottles by adding ultrapure HCl. The bottles were thoroughly rinsed with the sample water
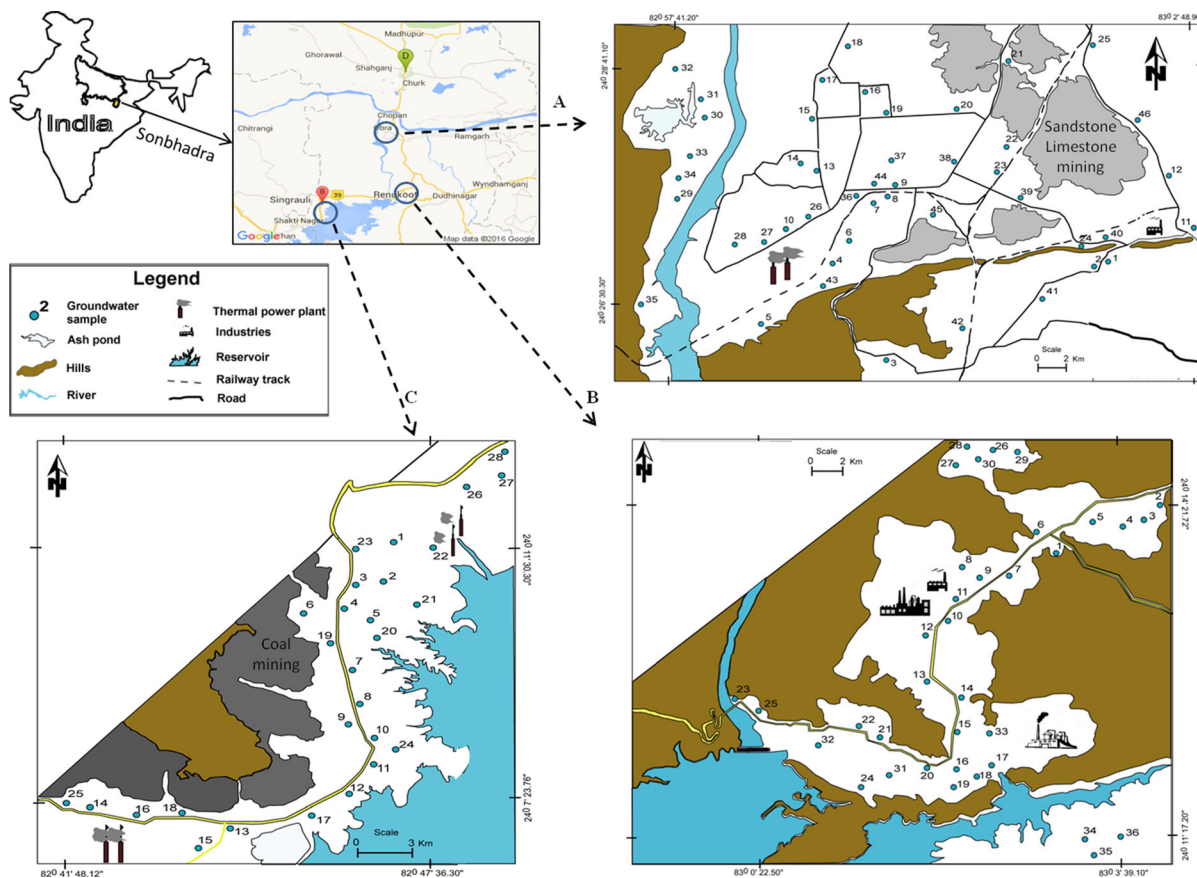
Fig. 1 Physiographic map of the three industrial clusters showing the sampling locations **A** Obra; **B** Renukoot; **C** Anpara

before filling and were then adequately labelled. Proper care was taken during sample collection, and the groundwater samples were collected only after 5–10 min of water flushing in order to stabilize its EC apart from removing the interference of standing water in the metal casing. Digital pH and EC meters were employed for recording the pH and EC, respectively, on site. The groundwater samples were then stored at 4 °C to avert any major chemical alteration. The solution was then filtered through 0.45 μm milli-pore filter paper prior to analysis.

Analysis and quality control and quality assurance

This study has used the atomic absorption spectrophotometer (Thermo Scientific M series AAS) for quantification of the various heavy metals (such as Cu, Fe, Zn, Cd, Co, Mn, Ni, Cr, Pb and As). The acidified arsenic samples were analysed total arsenic using the hydride generation method in atomic absorption

spectrometry. For the purpose of validation and cross-checking of results, at every 10th sampling location, a duplicate sample from the field was collected. The study used analytical grade chemicals of 1000 mg/l certified standard solutions (Merck, Darmstadt, Germany) for the purpose of sample preparation and its analysis. For each element, a standard calibration curve was prepared by employing the serial dilution of corresponding standard solutions of metal ions.

For performing mercury analysis of the groundwater samples, the Atomic Fluorescence Spectrophotometer (AFS) (instrument model AF-420, PG Instruments) that was equipped with an auto-sampler, high-intensity hollow cathode lamp and hydride generator assembly was used. For the purpose of data integration and processing, the manufacturer brand PF Win software was employed. Also the instrument was set at 50 mA lamp current, carrier gas flow of around 1 ml/min, scanning time of 15 s. Sample was

atomized at 100 °C with negative voltage of 200 V. Under the optimized spectroscopic condition, a steady baseline was obtained. Dilutions, if any (in case of sample mercury concentrations greater than the highest calibrated standard value), were made using 10% HCl only. In a 15-ml centrifuge tube, a 0.1 ml 1000 ppb stock solution was taken with 1.9 ml of 10% HCl. In order to give a concentration of 10 ppb, the mix was diluted up to 10 ml with 10% HCl solution. Further, the calibration curve within the range of 0.5–5 ng/ml was plotted in order to calculate the regression coefficient, slope and intercept. For each heavy metal, the R^2 value of the calibration curve was observed to be > 0.99. The study used double-distilled water in consideration of the data quality assurance throughout the analysis, wherein the reproducibility of the analytical data came to be under 5% whereas the analytical error was estimated to be $< 10\%$.

Daily consumption of drinking water

A random questionnaire survey was carried out in the study region to collect information about the daily intake of the drinking water by the local residents. For this study, only those residents were selected who mainly drink water from borewells/hand pumps present in the study area. It surveyed consisted of 178 adults (117 male and 61 female) aged between 18 and 70 years and 90 children aged between 11 and 17 years. Adults were selected based on their different walks of lifestyle such as home dwellers, office going people, shopkeepers, labourers, mining workers and farmers. For children, daily water intake data were collected from different schools situated in the study region. They were instructed to check how much amount of water is consumed in a day in a particular vessel and how many times. The study used two calibrated glasses, each with the capacity of 250 ml and 500 ml, in order to estimate the correct volume of the daily water consumption by the respondents. These calibrated glasses were confirmed with the correct amount and frequency of the respondent's drinking water intake from their vessels. These data were converted into litres and then averaged to calculate individual daily intake.

Heavy metal pollution index (HPI)

The HPI is a standard quality measure employed for assessing heavy metals presence in the water. Weighted arithmetic quality mean is used to compute this index through two steps. Firstly, a rating scale is prepared, wherein the selected heavy metals are assigned weight. Secondly, pollution parameters are selected to compute pollution index of the area. The established rating scale provides any arbitrary value from 0 to 1 depending upon the significance of the individual parameters in affecting the overall quality. In the present study, a unit weight (W_i) is considered as a value that is inversely proportional to the recommended standard (S_i) of the corresponding parameter (i.e. Fe, Mn, Co, Cu, Cd, Ni, Pb, Zn, Cr, As and Hg). These standards are in accordance with the WHO 2011 drinking water standards. The values of HPI model are calculated as per Eq. (1) (Prasad et al. 2014).

$$\text{HPI} = \frac{\sum_{i=1}^{n} W_i Q_i}{\sum_{i=1}^{n} W_i}. \tag{1}$$

Here, W_i represents the unit weight of the ith parameter, n denotes the number of parameters taken into consideration, whereas the Q_i signifies the subindex of the ith parameter, which is calculated as per Eq. (2).

$$Q_i = \sum_{i=0}^{n} \{M_i(-)I_i\}/(S_i - I_i) \times 100 \tag{2}$$

where M_i characterizes the value examined for the heavy metal for its ith parameter (analytical data), I_i denotes ith parameter's ideal value, and finally, S_i signifies ith parameter's standard value.

In the above stated equation, $(-)$ sign sans any algebraic importance as it specifies the numerical difference between the values. The HPI model employed in the current study intends to evaluate suitability of water for drinking and so its critical pollution index is set at 100.

Human health risk assessment

There are a number of ways through which a person can ingest heavy metals, but compared to oral routes (through food and water), intake via inhalation and

dermal absorption is negligible (O'Rourke et al. 1999; Muhammad et al. 2011; Khan et al. 2013a, b; Ahamad et al. 2018b). Exposure to the heavy metals causes human health risk. Non-carcinogenic risks can be examined through THQ and HI. The THQ is based on the ratio between an estimated dose of contaminant and the reference dose, below which there is no appreciable risk (Storelli 2008). In order to calculate the THQ through water ingestion, the US Environmental Protection Agency (USEPA 1989; Pawar and Pawar 2016) equation (Eq. 3) is used. It is as follows:

$$THQ = (EFr \times ED_{tot} \times SFI \times MCS_{inorg})/(RfD \times BWa \times ATn). \quad (3)$$

Here, EFr denotes exposure frequency (350 days/year); ED signifies exposure duration {58 years for adults and 10 years for children (Chai et al. 2010)}; SFI represents water ingestion rate (2.83 l for adults and 2.18 litre for children, as per our questionnaire survey); MCS_{inorg} characterizes metal concentration in water (μg/l); BWa indicates average body weight {which is 70 kg for adult and 20 kg for children (USEPA 2002)}; ATn specifies the time period over which the dose is averaged (365 days/year $\times ED_{tot}$ for non-carcinogen) and 70 (lifetime year) $\times$ 365 days for carcinogen (USEPA 2002) and RfD represents the oral reference dose (in μg/kg/day). The RfD values for Zn, Cd, Fe, Pb, Mn, Cu, Cr, Ni, As and Hg are 300, 0.5, 300, 4, 20, 40, 3, 20, 0.3 and 0.3, respectively (USEPA 2006; Wu et al. 2009; Gupta et al. 2015). The HI is used to describe the cumulative non-carcinogenic effect for more than one element and is calculated as follows:

$$HI = \sum_{i=1}^{n} HQ. \quad (4)$$

If the THQ and HI value is less than 1, then the non-carcinogenic toxic risk is regarded to be low. As it fluctuates beyond 1, there is a possibility of occurrence of a potential health risks.

According to USEPA, heavy metals such as Pb, Cr and As are considered as carcinogenic for human. Hence, carcinogenic risk has been calculated for these metals by using the given equation (Eq. 5):

$$Cancer\ risk(CR) = Chronic\ Daily\ Intake\ (CDI) \times Slope\ Factor. \quad (5)$$

Here, slope factor is in μg/kg/day. Slope factor for Pb, Cr and As is 8.5, 500 and 1500, respectively (USDOE 2011; USEPA 2005). Calculated value of cancer risk signifies the probabilities of an individual to develop cancer by the exposure of carcinogens for their lifetime. The acceptable or tolerable CR is $1 \times 10^{-6} - 1 \times 10^{-4}$.

Result and discussion

pH, EC and heavy metals in groundwater

Table 1 shows the statistical summary of pH, EC and concentrations of different heavy metals and their comparisons with WHO (2011) and BIS (2012) standards. The pH value showed that groundwater samples in the area under study were slightly acidic to alkaline in nature but within the permissible limit as per the standards set by the WHO and BIS. The EC value highlighted that majority of the groundwater samples from across the three clusters were within the WHO permissible ranges. The seasonal variation observed in the EC may be due to leaching of major cations and anions of salts from the soil or dissolution of minerals from the walls of aquifers because of rise in the water table (Kumar et al. 2009).

The concentration of Fe in groundwater samples exceeded by 74, 78 and 93% in postmonsoon and 70, 81 and 93% in premonsoon beyond the permissible limit prescribed by WHO (2011) and BIS (2012) in Obra, Renukoot and Anpara industrial clusters, respectively. Natural (iron containing minerals) and anthropogenic both sources may be responsible for the high content of Fe in the whole study region. The consumption of water with the high concentration of iron for long periods could affect the liver and consequently cause hemosiderosis disease (Bhaskar et al. 2010; Khan et al. 2013b). Mn value in groundwater samples exceeded by 41, 72 and 25% in postmonsoon and 41, 64 and 32% in premonsoon from the permissible limit prescribed by WHO (2011) in Obra, Renukoot and Anpara industrial clusters, respectively. The Cu concentration in the groundwater samples across the study area was all within the WHO and BIS standards. The value of Cd in the groundwater samples exceeded by 83, 89 and 97% in postmonsoon and 87, 89 and 100% in premonsoon beyond the

Table 1 Statistical summary of heavy metal value in groundwater samples of southern Sonbhadra around industrial cluster and its comparison with WHO and BIS standards

Element	WHO (2011)	BIS (2012)	Postmonsoon					Premonsoon				
			Min	Max	Mean	% samples exceeding WHO	% samples exceeding BIS limit	Min	Max	Mean	% samples exceeding WHO	% samples exceeding BIS limit
Obra (N = 46)												
pH	9	8.5	5.54	7.49	7.03	–	–	5.46	7.46	7.07	–	–
EC	1500	–	308.96	2121.91	1193.37	17	–	377.27	1880.14	1074.89	9	–
Fe	300	300	24.5	38,208.5	2470.37	74	74	19.8	43,540.3	2725.89	70	70
Mn	100	300	18.6	1699.1	209.18	41	24	23.1	2458.6	227.82	41	20
Cu	2000	1500	32.4	108.9	82.26	–	–	21.8	116	81.99	–	–
Cd	3	3	0.1	119	22.06	83	83	0.1	55.3	23.19	87	87
Ni	70	20	0.1	72	23.12	4	54	0.1	79	25.99	4	54
Zn	4000	15,000	0.1	2363.3	452.99	–	–	0.1	2784.7	511.24	–	–
Cr	50	50	0.1	975	90.57	20	20	0.1	867	59.96	28	28
Co	–	–	12.4	74.4	48.34	–	–	9.5	79	47.99	–	–
Pb	10	10	0.1	110	36.17	89	89	0.1	94	38.75	91	91
As	10	50	1.11	19.4	6.48	15	–	0.59	20.4	5.64	17	–
Hg	1	1	0.54	3.86	2.01	85	85	0.42	4.12	2.14	89	89
Renukoot (N = 36)												
pH	9	8.5	6.3	8	7.1	–	–	6.6	8.1	7.15	–	–
EC	1500	–	373.34	1857.14	901.76	11	–	340.86	1775.13	849.79	6	–
Fe	300	300	125	14,910	1583.14	78	78	116.1	15,071.5	1589.35	81	81
Mn	100	300	40	1140	278.54	72	28	48.7	1188	255.55	64	28
Cu	2000	1500	66	996	169.02	–	–	73.9	985	150.32	–	–
Cd	3	3	0.1	189	28.44	89	89	0.1	279	35.94	89	89
Ni	70	20	0.1	83.1	31.94	14	61	0.1	91.4	40.73	25	72
Zn	4000	15,000	8.9	12,179.4	1675.52	17	–	2.4	14,680	1947.8	19	–
Cr	50	50	0.1	975	138.84	36	36	0.1	994	105.47	53	53
Co	–	–	3	269	32.06	–	–	5	314	58.11	–	–
Pb	10	10	0.1	104	28.96	69	69	0.1	127	36.38	69	69
As	10	50	1.5	20.3	9.23	39	–	1.11	19.6	8.76	39	–
Hg	1	1	0.57	15.68	2.29	86	86	0.68	15.21	2.37	86	86

Table 1 continued

Element	WHO (2011)	BIS (2012)	Postmonsoon					Premonsoon				
			Min	Max	Mean	% samples exceeding WHO	% samples exceeding BIS limit	Min	Max	Mean	% samples exceeding WHO	% samples exceeding BIS limit
Anpara (N = 28)												
pH	9	8.5	7.04	7.8	7.36	–	–	6.93	8	7.31	–	–
EC	1500	–	561.67	1531.75	809.96	4	–	654.4	1645.02	892.26	4	–
Fe	300	300	47.2	9568.8	2227.44	93	93	84	9421	2352.86	93	93
Mn	100	300	21	1169	176.07	25	11	32	1384	209.14	32	11
Cu	2000	1500	14	975	124.11	–	–	27	314	105.86	–	–
Cd	3	3	1.4	97.8	17.53	97	97	3.8	112	21.8	100	100
Ni	70	20	8	1140	166.79	18	82	18	1280	164.96	21	93
Zn	4000	15,000	2.6	3254.5	605.25	–	–	11	3512	720.61	–	–
Cr	50	50	15.1	148.7	50.01	25	25	34.2	189	65.98	54	54
Co	–	–	1.8	48	16.83	–	–	11	57	29.79	–	–
Pb	10	10	7	540	102	93	93	18	780	132.68	100	100
As	10	50	4.3	20	10.81	61	–	6.54	18.32	11	53	–
Hg	1	1	1.64	11.14	4.9	100	100	1.81	10.25	4.98	100	100

All metals values are in ppb (µg/l) except pH and EC (µS/cm) at 25 °C

permissible limit prescribed by the WHO and BIS in Obra, Renukoot and Anpara industrial clusters, respectively. The contaminated water with high values of the Cd has the potential to cause various medical complications like renal failure, muscle cramps, nausea and liver injury (Alam et al. 2012). The concentration of Ni in groundwater samples exceeded by 4, 14 and 18% in postmonsoon and 4, 25 and 21% in premonsoon from the permissible limit prescribed by WHO (2011) in Obra, Renukoot and Anpara industrial clusters, respectively. The value of Ni in groundwater samples exceeded by 54, 61 and 82% in postmonsoon and 54, 72 and 93% in premonsoon beyond the permissible limit prescribed by BIS (2012) in Obra, Renukoot and Anpara industrial cluster, respectively. The drinking water with high concentration of nickel could result in many medical problems like birth defects, heart disorders and respiratory failures (Khan et al. 2013b).The concentration of Cr in groundwater samples was found to exceed by 20, 36 and 25% in postmonsoon and 28, 53 and 54% in premonsoon from the permissible limit prescribed by WHO and BIS in Obra, Renukoot and Anpara industrial clusters, respectively. Ingestion of high amounts of Cr in the drinking water can result in the lung cancer and allergic incidents (Puri et al. 2011). The value of Pb in groundwater samples exceeded by 89, 69 and 93% in postmonsoon and 91, 69 and 100% in Premonsoon from the permissible limit prescribed by WHO and BIS in Obra, Renukoot and Anpara industrial cluster, respectively. High Pb concentration in drinking water samples can cause blood disorder, brain problems along with mutilate nervous connections (Mohod and Dhote 2013). Exposure to Pb may lead to carcinogenic effects (AbRazak et al. 2016). The value of As in groundwater samples was found to exceed by 15, 39 and 61% in postmonsoon and 17, 39 and 53% in premonsoon beyond the permissible limit prescribed by WHO (2011) in Obra, Renukoot and Anpara industrial clusters, respectively. As concentration in all the clusters highlighted that overall groundwater samples were under the permissible limit prescribed by BIS. Ingestion of As rich water results in hyperpigmentation and keratosis and ultimately skin cancer (Kumar et al. 2010). The mean values of Hg in the groundwater samples were 2.01 µg/l, 2.29 µg/l and 4.90 µg/l in postmonsoon and 2.14 µg/l, 2.37 µg/l and 4.98 µg/l in premonsoon in Obra, Renukoot and Anpara industrial clusters, respectively. The

concentration of Hg in groundwater samples exceeded by 85, 86 and 100% in postmonsoon and 89, 86 and 100% in premonsoon from the permissible limit (1 µg/l) prescribed by WHO and BIS in Obra, Renukoot and Anpara industrial clusters, respectively. High concentrations of Hg may lead to adverse effects on brain, liver, nervous system, gingivitis tremors and heart muscle (Sun et al. 2013; Nazarpour et al. 2018). Earlier studies of heavy metals in groundwater samples from coal mining and thermal power plant region of Singrauli have also reported high concentrations of Cd, Cr, Pb, As, Fe, Ni (Agrawal et al. 2011; Khan et al. 2013a; Rai et al. 2015).

Heavy metal pollution index (HPI)

The HPI is considered an important valuable tool in measuring the pollution load as well as determining composite influence of metals over the quality of the water (Sheykhi and Moore 2012). HPI value was calculated for the three clusters for both postmonsoon and premonsoon seasons. It was found that HPI values of every cluster were higher than the suggested critical value of the pollution index (100). The highest HPI value was 510.26 and 565.2 for Anpara (cluster C), followed by Renukoot (Cluster B) 374 and 435, and Obra (Cluster A) 313 and 330, in the postmonsoon and premonsoon seasons, respectively (Table 2). In terms of seasonal variation of pollution index, it was lower in postmonsoon in comparison to premonsoon in all the clusters which may be the effect of dilution. The pollution level in the study region is intolerable during both the seasons. Arslan et al. (2017) found very high value of HPI (360) and (440) for dry season and wet seasons, respectively. Prasad et al. (2014) found similar trend of HPI, high for premonsoon (6.2) and low for postmonsoon (3.2). As per Table 2 and Fig. 2, HPI values were more than 100 (critical value) for other two clusters except Obra in which only two samples, one in the postmonsoon and another in the premonsoon showed HPI value less than 100. The highest value of HPI was found for Renukoot cluster (Fig. 2). Overall, the heavy metal pollution load in the study region is very high during both the seasons.

Table 2 Statistical summary of heavy metal pollution index (HPI) of all the three industrial clusters in southern Sonbhadra

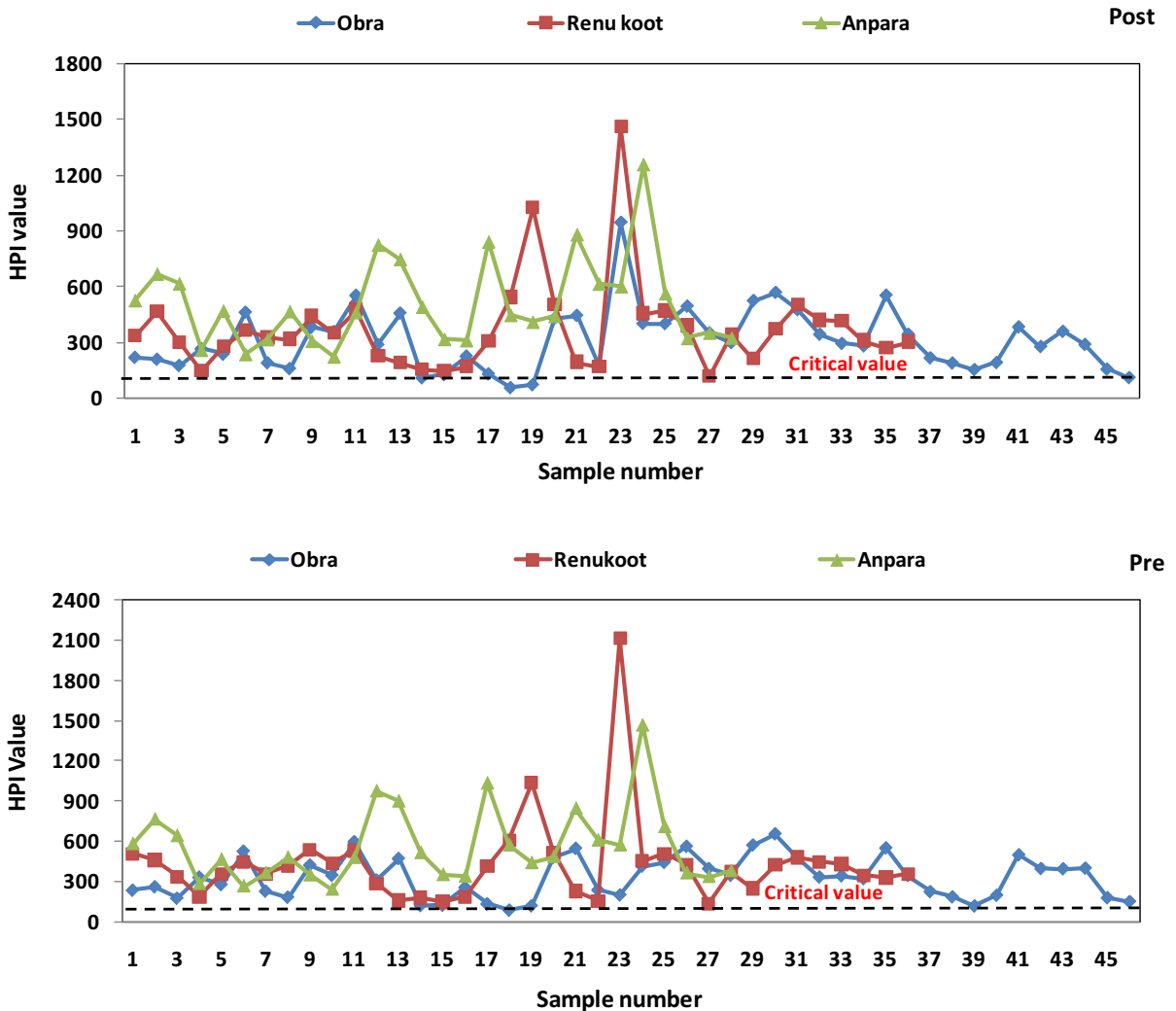|	Obra		Renukoot		Anpara	
	Post	Pre	Post	Pre	Post	Pre
Min	59.48	87.97	118.80	138.80	223.22	242.93
Max	945.98	654.66	1459.92	2117.24	1256.51	1466.11
Average	313.79	330.46	374.04	435.26	510.26	565.20

Fig. 2 Seasonal variation of HPI for each sampling location for all the three clusters in postmonsoon and premonsoon season

Human health risk assessment

Non-carcinogenic risk (NCR)

The metal accumulation in the groundwater resources because of the widespread urbanization and industrialization in the study area is likely to have a direct impact on both the human health as well as the ecosystem since the local population is primarily dependent on the groundwater for drinking purposes. THQ and HI were estimated to see the non-carcinogenic risks in the study region.

Target hazard quotient (THQ)

Table 3 summarizes the THQ values of the studied metals in the groundwater samples collected from the industrial clusters of southern Sonbhadra. For adults and children, the study found the THQ values of heavy metals for the Cluster A (Obra) were in the following order: Cd > Cr > As > Mn > Pb > Fe > Hg > Cu > Zn > Ni during both seasons. Likewise for adults and children in Cluster B (Renukoot), the THQ values of heavy metals were in order Cd > Cr > As > Mn > Hg > Pb > Zn > Fe > Cu > Ni   and Cd > Cr > As > Mn > Pb > Hg > Zn > Fe > Cu > Ni, respectively, during postmonsoon and premonsoon seasons. For adults and children in Cluster C (Anpara), the THQ values of heavy metals were in the order   As > Cd > Pb > Cr > Hg > Mn > Ni > Fe > Cu > Zn   and   Cd > As > Pb > Cr > Hg > Mn > Ni > Fe > Cu > Zn in postmonsoon and premonsoon season, respectively. THQ value of Cd for adults exceeded the safe limit by 50 and 57% for Obra, by 67 and 81% for Renukoot and by 29 and 39% for Anpara, during postmonsoon and premonsoon, respectively. The THQ value of Cr was exceeding the safe limit by 13 and 17% for Obra, by 28 and 36% for Renukoot and by 14 and 21% for Anpara cluster in postmonsoon and premonsoon, respectively, for adults. For adults, THQ values of As exceeded the safe limit by 33 and 24% for Obra, by 58 and 44% for Renukoot and by 75 and 75% for Anpara, in postmonsoon and premonsoon, respectively. THQ value of Cd exceeded the safe limit by 78 and 83% for Obra, by 83 and 86% for Renukoot and by 96 and 96% for Anpara, in postmonsoon and premonsoon, respectively, for children. THQ values of Cr were found to exceed the safe limit by 57 and 59% for Obra, by 57

and 57% for Renukoot and by 82 and 100% for Anpara, in postmonsoon and premonsoon, respectively, for children. For children, Pb THQ values exceeded the safe limit by 39 and 43% for Obra, by 28 and 39% for Renukoot and by 71 and 75% for Anpara in postmonsoon and premonsoon, respectively. THQ value of Hg for children exceeds the safe limit by 13 and 17% for Obra, by 14 and 14% for Renukoot and by 86 and 82% for Anpara during the postmonsoon and premonsoon, respectively. THQ values for As exceeded the safe limit by 65 and 59% for Obra, by 89 and 92% for Renukoot and by 100 and 100% for Anpara cluster, in postmonsoon and premonsoon, respectively, for children. If the groundwater from the borewells with high THQ value (> 1) for the observed metals is used for a longer and sustained periods without adopting any remedial measures, these will have the potential to cause NCR human health risks to the consumers in the region. In the majority of the groundwater samples, the results highlighted a significant non-cancer health hazard across the study area as indicated by the presence of more than unity THQ values for Cr, Cd, As, Pb and Hg. Since the heavy metals are extremely persistent (non-biodegradable and non-thermodegradable) and readily increase to the toxic levels in the environment (Sharma et al. 2007), the baselines are required for assessing the current status of the environment (Galan et al. 2008). Within this context, the current study assists in establishing a periodic monitoring network in order to evaluate the trace elements contamination that reveals both the quality of groundwater as well as the level of associated health risk.

Hazard index (HI)

The HI is used to describe the cumulative non-carcinogenic effect of all the studied metals. Table 3 shows the mean values of HI and their seasonal variation for adult and children in all the three clusters. The mean value of HI for adults in the groundwater samples was 5.24, 6.96 and 6.18 in postmonsoon and 4.95, 7.12 and 7.13 in premonsoon, whereas for children it was 14.18, 18.84 and 16.72 in postmonsoon and 13.39, 19.28 and 19.28 in premonsoon, in Obra, Renukoot and Anpara, respectively. For other two clusters, HI values decreased in postmonsoon as compared to premonsoon, except Obra, where it was found to increase. This may be due to high leaching of

Table 3 Statistical summary of target hazard quotient (THQs) and hazard index (HI) for adult and children through ingestion pathway in southern Sonbhadra

Parameter	Obra ($N = 46$)				Renukoot ($N = 36$)				Anpara ($N = 28$)			
	THQ_{adult}		$THQ_{children}$		THQ_{adult}		$THQ_{children}$		THQ_{adult}		$THQ_{children}$	
	Post	Pre	Post	Pre	Post	Pre	Post	Pre	Post	Pre	Post	Pre
Fe												
Min	0.003	0.003	0.009	0.007	0.016	0.015	0.044	0.041	0.006	0.011	0.017	0.029
Max	4.942	5.631	13.373	15.239	1.928	1.949	5.219	5.275	1.238	1.218	3.349	3.297
Mean	0.320	0.353	0.865	0.954	0.205	0.206	0.554	0.556	0.288	0.304	0.780	0.824
% samples exceeding	7	7	7	9	6	6	8	6	4	4	29	32
Mn												
Min	0.036	0.045	0.098	0.121	0.078	0.094	0.210	0.256	0.041	0.062	0.110	0.168
Max	3.296	4.770	8.920	12.908	2.212	2.305	5.985	6.237	2.268	2.685	6.137	7.266
Mean	0.406	0.442	1.098	1.196	0.540	0.496	1.462	1.342	0.342	0.406	0.924	1.098
% samples exceeding	11	9	30	30	17	11	58	42	11	14	18	18
Cu												
Min	0.031	0.021	0.085	0.057	0.064	0.072	0.173	0.194	0.014	0.026	0.037	0.071
Max	0.106	0.113	0.286	0.305	0.966	0.955	2.615	2.586	0.946	0.305	2.559	0.824
Mean	0.080	0.080	0.216	0.215	0.164	0.146	0.444	0.395	0.120	0.103	0.326	0.278
% samples exceeding	–	–	–	–	–	–	8	6	–	–	4	–
Cd												
Min	0.008	0.008	0.021	0.021	0.008	0.008	0.021	0.021	0.109	0.295	0.294	0.798
Max	9.234	4.291	24.990	11.613	14.666	21.650	39.690	58.590	7.589	8.691	20.538	23.520
Mean	1.712	1.800	4.633	4.870	2.207	2.789	5.973	7.548	1.360	1.692	3.680	4.579
% samples exceeding	50	57	78	83	67	81	83	86	29	39	96	96
Ni												
Min	0.000	0.000	0.001	0.001	0.000	0.000	0.001	0.001	0.016	0.035	0.042	0.095
Max	0.140	0.153	0.378	0.415	0.161	0.177	0.436	0.480	2.212	2.483	5.985	6.720
Mean	0.045	0.050	0.121	0.136	0.062	0.079	0.168	0.214	0.324	0.320	0.876	0.866
% samples exceeding	–	–	–	–	–	–	–	–	14	11	14	11
Zn												
Min	0.000	0.000	0.000	0.000	0.001	0.000	0.003	0.001	0.000	0.001	0.001	0.004
Max	0.306	0.360	0.827	0.975	1.575	1.899	4.263	5.138	0.421	0.454	1.139	1.229
Mean	0.059	0.066	0.159	0.179	0.217	0.252	0.586	0.682	0.078	0.093	0.212	0.252
% samples exceeding	–	–	–	–	6	6	19	17	–	–	4	4
Cr												
Min	0.001	0.001	0.004	0.004	0.001	0.001	0.004	0.004	0.195	0.442	0.529	1.197
Max	12.610	11.213	34.125	30.345	12.610	12.856	34.125	34.790	1.923	2.444	5.205	6.615
Mean	1.171	0.775	3.170	2.099	1.796	1.364	4.859	3.691	0.647	0.853	1.750	2.309
% samples exceeding	13	17	57	59	28	36	57	57	14	21	82	100
Pb												
Min	0.001	0.001	0.003	0.003	0.001	0.001	0.003	0.003	0.068	0.175	0.184	0.473
Max	1.067	0.912	2.888	2.468	1.009	1.232	2.730	3.334	5.238	7.566	14.175	20.475
Mean	0.351	0.376	0.949	1.017	0.281	0.353	0.760	0.955	0.989	1.287	2.678	3.483
% samples exceeding	2	–	39	43	3	6	28	39	11	14	71	75
As												

Table 3 continued

Parameter	Obra (N = 46)				Renukoot (N = 36)				Anpara (N = 28)			
	THQ_{adult}		$THQ_{children}$		THQ_{adult}		$THQ_{children}$		THQ_{adult}		$THQ_{children}$	
	Post	Pre	Post	Pre	Post	Pre	Post	Pre	Post	Pre	Post	Pre
Min	0.144	0.076	0.389	0.207	0.194	0.144	0.525	0.389	0.556	0.846	1.505	2.289
Max	2.509	2.638	6.790	7.140	2.625	2.535	7.105	6.860	2.587	2.369	7.000	6.412
Mean	0.838	0.730	2.269	1.975	1.194	1.133	3.231	3.066	1.398	1.423	3.783	3.852
% samples exceeding	33	24	65	59	58	44	89	92	75	75	100	100
Hg												
Min	0.070	0.054	0.189	0.147	0.074	0.088	0.200	0.238	0.212	0.234	0.574	0.634
Max	0.499	0.533	1.351	1.442	2.028	1.967	5.488	5.324	1.441	1.325	3.899	3.586
Mean	0.260	0.277	0.704	0.749	0.296	0.307	0.800	0.830	0.634	0.644	1.716	1.743
% samples exceeding	–	–	13	17	3	3	14	14	18	14	86	82

Statistics	HI_{adult}		$HI_{children}$		HI_{adult}		$HI_{children}$		HI_{adult}		$HI_{children}$	
	Post	Pre	Post	Pre	Post	Pre	Post	Pre	Post	Pre	Post	Pre
Min	1.01	1.48	2.74	4.02	1.77	2.00	4.80	5.43	3.08	3.78	8.32	10.23
Max	17.80	18.84	48.17	51.00	21.23	29.47	57.45	79.75	21.41	25.54	57.93	69.11
Mean	5.24	4.95	14.18	13.39	6.96	7.12	18.84	19.28	6.18	7.13	16.72	19.28

metals from the surface with rain water. Radar plot of the HI of all samples in all the three clusters is shown in Fig. 3(I, II) for adult and Fig. 3(III, IV) for children during postmonsoon and premonsoon. It was observed that all samples of the three clusters showed more than one value of the safe limit of HI. The HI value for children was observed to be very high compared to adults, which means that children within the study area are more prone to non-carcinogenic health risk.

Carcinogenic risk (CR)

The cancer slope risk factor is only available for Pb, As and Cr; therefore, carcinogenic risk was observed for these elements in adults and children living around these industrial clusters. The mean value of CR for Cr in case of adults in the groundwater samples varied from 1455 to 963 in Obra, 2230–1694 in Renukoot and 803–1060 in Anpara during postmonsoon and premonsoon, respectively (Table 4). The mean value of CR for Cr in case of children varied from 676 to 448 in Obra, 1036–787 in Renukoot and 373–493 in Anpara during postmonsoon and premonsoon, respectively. The mean value of CR for Pb in case of adults in the groundwater samples during post- and premonsoon in Obra, Renukoot and Anpara regions varied between 9.87–10.6, 7.91–9.93 and 27.9–36.2, respectively. The

mean value of CR for Pb in case of children during post- and premonsoon in Obra, Renukoot and Anpara varied between 4.59–4.92, 3.68–4.62 and 12.94–16.84, respectively. The mean value of CR for As during post- and premonsoon varied between 312–272, 445–422 and 521–530 for adults in groundwater samples of Obra, Renukoot and Anpara region, respectively. In Obra, Renukoot and Anpara clusters, for children, the mean value of CR for As varied between 145–126, 207–196 and 242–247 during the post- and premonsoon, respectively.

Overall, carcinogenic risk due to Cr was found highest in the study region followed by As and Pb. Renukoot (Cluster B) showed highest carcinogenic risk due to Cr followed by Obra (Cluster A) and Anpara (Cluster C) for both adults and children. Anpara (cluster C) displayed highest carcinogenic risk of Pb followed by Obra (Cluster A) and Renukoot (Cluster B) for both adult and children. Anpara (cluster C) showed highest carcinogenic risk due to As followed by Renukoot (Cluster B) and Obra (Cluster A) in case of both adult and children. Carcinogenic Risk is higher for adult than children in the entire region. The CR for Cr, Pb and As was found very high than the tolerable limit of 1×10^{-4} – 1×10^{-6} for cancer risk. Thus, it may induce carcinogenesis in the
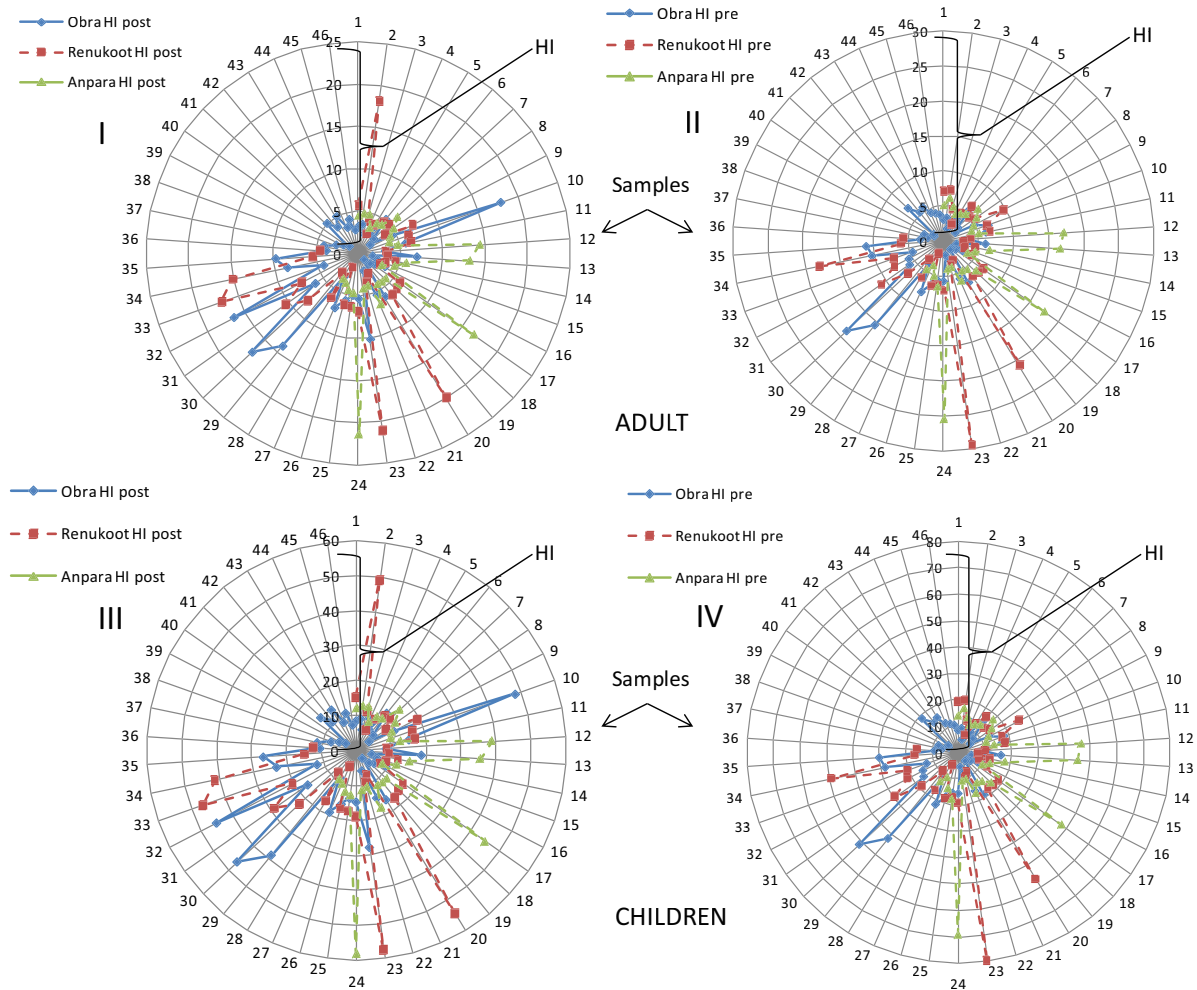
Fig. 3 Radar plot for hazard index (HI) of each sampling location during post- and premonsoon in the study area (**I**, **II**) for adults (**III**, **IV**) for children via consumption of drinking water

body of the residents. CR was higher for adults than children in all the three clusters (Table 4).

Carcinogenic risk mapping of As, Cr and Pb in three clusters during post- and premonsoon is shown in Supplementary Fig. 1 (1A to 1C, 2A to 2C and 3A to 3C) and Fig. 2 (4A to 4C, 5A to 5C and 6A to 6C) for adults and children, respectively. Carcinogenic risk map of As during postmonsoon and premonsoon season for adults and children revealed that highest cancer risk was found in groundwater samples very near to fly ash pond in western part in Obra; in southwest and south-eastern part in Renukoot, which are in proximity to chlor-alkali and aluminium smelting plants; in central and south-western part of Anpara, very near to coal mining and a thermal power plant.

Similar results were obtained for carcinogenic risk map of Cr for both seasons and demographics. Carcinogenic risk map of Pb during postmonsoon and premonsoon season for both adults and children revealed that highest cancer risk was found in central part of Obra which is very near to thermal power plant; in northern part of Renukoot which is in proximity to hi-tech carbon plant; in southern portion of Anpara which is near to a thermal power plant.

Although these findings are significant, the result may suffer from uncertainties since only daily water intake was measured for adult and children in the study area. In this study, only that water which is consumed for drinking is considered but other sources of intake such as tea, coffee, milk, juice and food. have not been

Table 4 Statistical summary of cancer risk (CR) for both adult and children in selected industrial clusters of southern Sonbhadra through ingestion pathway

Parameter	Obra				Renukoot				Anpara			
	CR_{adult}		$CR_{children}$		CR_{adult}		$CR_{children}$		CR_{adult}		$CR_{children}$	
	Post	Pre	Post	Pre	Post	Pre	Post	Pre	Post	Pre	Post	Pre
Cr												
Min	1.606	1.606	0.747	0.747	1.606	1.606	0.747	0.747	242.506	549.252	112.722	255.303
Max	15,658.500	13,924.020	7278.375	6472.155	15,658.500	15,963.640	7278.375	7420.210	2388.122	3035.340	1110.046	1410.885
Mean	1454.547	962.972	676.102	447.608	2229.708	1693.795	1036.412	787.309	803.115	1059.673	373.303	492.557
Pb												
Min	0.027	0.027	0.013	0.013	0.027	0.027	0.013	0.013	1.911	4.914	0.888	2.284
Max	30.032	25.664	13.960	11.929	28.394	34.674	13.198	16.117	147.431	212.956	68.529	98.986
Mean	9.874	10.580	4.590	4.918	7.907	9.933	3.675	4.617	27.848	36.224	12.944	16.838
As												
Min	53.480	28.426	24.858	13.213	72.270	53.480	33.593	24.858	207.174	315.097	96.299	146.463
Max	934.692	982.872	434.463	456.858	978.054	944.328	454.619	438.942	963.600	882.658	447.900	410.276
Mean	312.311	271.855	145.168	126.363	444.728	422.030	206.718	196.168	520.688	530.217	242.026	246.455

considered. Additionally, other parameters such as reference dose, slope factor, exposure duration, body weight and exposure frequency were adopted from USEPA and USDOE standards, which may vary with a particular region's climatic condition. Despite these ambiguities, the findings of this study are meaningful and draw attention of the decision makers and can facilitate them in better management of industrial emissions and disposal of contaminated wastewater into land or water bodies, thereby, minimizing health risks to the inhabitants. Results of this study may be used as a database for current groundwater drinking standards which need revision for tropical countries due to high water intake rates there.

Conclusion

The concentration of the heavy metals and its comparison with the WHO and BIS standards showed that the groundwater is severely contaminated with Fe, Pb, Cd and Hg in Obra, with Fe, Mn, Pb, Cd and Hg in Renukoot and with Fe, Hg, As, Cd, Cr and Pb in Anapra during both the seasons. The HPI values of all the three clusters in both the seasons were higher than the critical value of 100. Postmonsoon value of HPI was lower in comparison to premonsoon in each cluster. High THQ values in majority of the samples suggested a substantial non-cancerous health hazard due to the presence of As, Cd, Cr, Hg and Pb in the study region. It was observed that all the samples in the three clusters exceeded the safe limit of HI, i.e. > 1. The HI value for children was found very high compared to adults indicating that children were more susceptible to non-carcinogenic health risk. Carcinogenic risk due to Cr, Pb and As was found very high as per safety guidelines of cancer risk. Carcinogenic risk due to Cr was observed highest in the study region followed by As and Pb. Based on the above analysis, it can be argued that for the region under study, the associated health risk due the metals could emerge as a severe threat in near future if the usage of groundwater continues for long term without adopting any proper remedial measures.

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