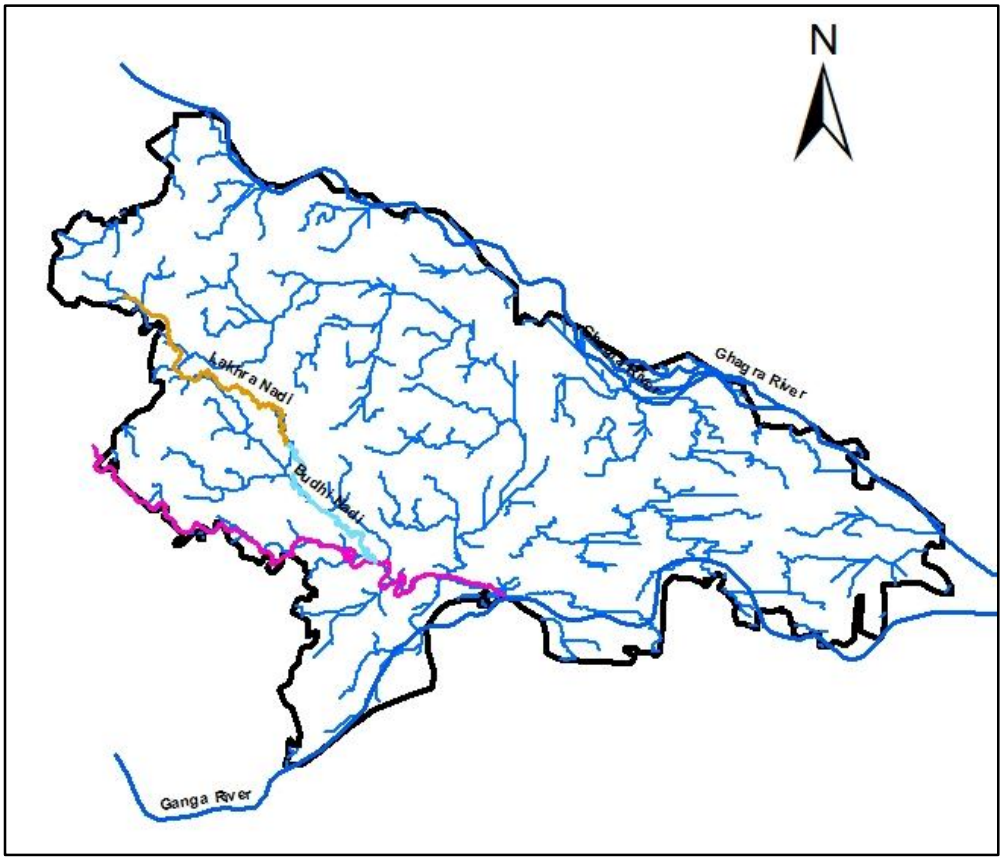
Alternate water supply management strategies in arsenic affected/vulnerable areas: Mapping of Arsenic affected zones/regions in Eastern U.P

Final Report



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PREFACE

Groundwater is an important and vital component of our life support system. The ground water resources are being utilized for drinking, irrigation and industrial purposes. There is growing concern on deterioration of ground water quality due to geogenic and anthropogenic activities. Ground water contamination with arsenic (As) is one of the major threats to human health. In India, high concentration of arsenic beyond permissible limit of 0.05 mg/l in ground water has been reported from 86 districts of 10 States. States like West Bengal, Bihar, Uttar Pradesh and Assam etc., are facing acute problem of arsenic contamination. The ground water of Eastern Uttar Pradesh has been reported to be severely contaminated with arsenic. Therefore, a study entitled "Alternate water supply management strategies in arsenic affected/vulnerable areas: Mapping of Arsenic affected zones/regions in Eastern U.P" was envisaged to identify and locate arsenic affected villages of Ballia district. Besides this, a spatial distribution map of arsenic was prepared to locate hotspot and vulnerable areas of groundwater contamination in the district.

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Abstract

Ground water is an important and vital component of our life support system. The ground water resources are being utilized for drinking, irrigation and industrial purposes. There is growing concern on deterioration of ground water quality due to geogenic and anthropogenic activities. Ground water contamination with arsenic (As) is one of the major threats to human health. The Ganges River basin (GRB), the water machine of India houses more than 8.3% of world population, with an area of 1.086 million km^2. Most part of the Gangetic basin (Uttar Pradesh (U.P.), Bihar and West Bengal) is extremely fertile for agricultural practices and have a very favorable condition for urbanization and industrialization but the basin has been reported with geogenic and anthropogenic contamination in surface and ground water. Elevated concentration of arsenic (As) in groundwater of the alluvial plains of GRB basin attracted maximum national and international attention. Groundwater quality in the states of U.P. indicates the presence of elevated concentrations of As in drinking water wells. One of the districts i.e. Ballia is severely affected by As contamination in groundwater. In the present study, Ballia district has been selected for the geo-chemical study with special reference to arsenic distribution mapping. Ballia is the eastern most district of Uttar Pradesh, covering an area of 2981 sq.km, lies in between 25º33' and 26º11'N latitudes and 83º 38' and 84º 39'E longitudes. The district is bounded on the north by Ghagra River and in the south by Ganga River. The study area has an average rainfall of 983 mm with mean annual temperature of 27ºC, which varies between 5.4ºC to 41.5ºC. Ground water samples were collected in pre-monsoon (May, 2016) season from India-marked-hand pump (110-140 ft depth) for all the seventeen administrative blocks of district by making grid of size 4 km x 4 km. The chemical analyses results revealed that calcium (Ca^{2+}) is found to be dominant cation in ground water samples followed by Na^+, Mg^{2+}, and K^+. The concentration of Ca^{2+} is found to be in the range of (21-142 mg/L, median 70 mg/L). The concentration of Na^+ and Mg^{2+} are varied in the range of (8.6-162 mg/L, median 31 mg/L) and (0.7-113 mg/L, median 34 mg/L) respectively. HCO_3^- is found to be major anion followed by Cl^- and $SO4^{--}$. The scatter diagram between (Ca^{2+}+Mg^{2+}) vs $Tz^+$for ground water showing a strongly positive correlation ((R^2=0.73), and reflects the high abundance of Ca^{2+}+Mg^{2+}) in the groundwater. The Piper diagram suggests that hydrogeochemical facies is of Ca–Mg-HCO3 type.

The chemical results shows that approximately ~58% of ground water samples collected from the study area are enriched with As concentrations >10 µg/L. Ground water with

elevated As (>100 μg/L) concentration is extensively present in the holocene aquifer of Shivpur Diar (245 μg/L), Pipra (141μg/L), Umraoganj (172 μg/L), Shivpur (168 μg/L), AsmanThola (181 μg/L), Siyarahiyan Village (229 μg/L), Bhawal Chapra (203 μg/L), Srinagar (219 μg/L) and Bhopalpur (187 μg/L). The maximum concentration of AS has been found to be 651 μg/L in the well of Ekauni (Dularampur) village of Sohaon administrative block of Ballia disctrict, which is close to the Ganges River. Arsenic showed a distinct relationship with several other groundwater solutes. Iron is positively correlated with As (0.612), but does not show any strong positive correlation with Mn (0.33). It is also observed that As has moderate positive correlation with SO_4^{2-} (0.572), which indicate that pyrite/sulphide oxidation may be one of the governing processes for As liberation/mobilization. The spatial distribution map of Arsenic indicates the region which are severely affected by As. These regions are located along the Ganga flood plains (typical hotspot near to convergences of Ghagra and Ganges river), generally in the south and northern east part of Ballia district.

1. Introduction

Arsenic is a metalloid with symbol As, has atomic number 33 and atomic mass 74.92. Arsenic forms colourless, odorless, crystalline compounds As_2O_3 and As_2O_5 which are hygroscopic in nature and readily soluble in water to form acidic solutions. The most common oxidation states for arsenic are:-3 (arsenide: usually alloy-like intermetallic compounds), +3 (arsenites (As (III)) and most organo-arsenic compounds) and +5 (arsenates (As(V)): the most stable inorganic arsenic oxy-compounds. It can exist in organic and inorganic form. Inorganic arsenic is generally more toxic than organic arsenic. Inorganic arsenic occurs naturally in many types of rocks and it is generally found with sulphide ore arsenopyrite. Inorganic arsenic compounds known to be human carcinogens. Arsenic in elemental form is insoluble in water but it is soluble in oxidised form. Arsenic is a natural constituent in bedrock and soil. It usually occurs at low concentrations (average 1–5 mg/kg) in the Earth's crust (Bhattacharya et al., 2002), but may be concentrated in certain rock types and especially in gold and sulphide-bearing ore deposits and occurrences. Pyrite [FeS_2; or arsenian pyrite $Fe(AsS)_2$] and arsenopyrite [FeAsS] are typical sulphide minerals containing As. These minerals are relatively stable in the bedrock and in deep soil under reducing and near-neutral conditions. However, the mobility of As is largely dependent on changes in the pH and redox conditions resulting from natural processes (e.g. microbial activity) or anthropogenic disturbance of earth materials (e.g. mining) (Smedley and Kinniburgh, 2002). The behaviour of As is also dependent on its oxidation state, As(III) and As(V) being the most commonly occurring inorganic species in the ground water environment (Smedley and Kinniburgh, 2002). As commonly precipitates as Fe(III) arsenates or adsorbs onto Fe(III)oxides and Fe(III)oxy-hydroxides.

Elevated concentrations of arsenic in drinking water (above 50 µg/L) have been reported in several countries, including Bangladesh, India, Argentina, Chile, China, Mongolia, Taiwan, Nepal, Japan, Mexico, Poland, Vietnam, and the USA.. Weathering processes of rocks and minerals appears to be a major source of arsenic found in soils. The arsenic concentration is usually higher in soils than in parent rocks, because it accumulates due to weathering and translocation in colloid fractions. Under typical soil forming conditions, the nature of arsenic in soil is controlled by the lithology of parent rock materials, volcanic activity, bioactivity, weathering history, transport, sorption, and precipitation. The concentrations of arsenic in non-contaminated soils range from 0.1 to 40 mg/kg. Peaty and peaty clay sediments at the depth of 7–10 m in southwest Bangladesh showed an arsenic

content of 20–111 mg/kg. Bedrock is the ultimate source of As, but eventually it moves into soil through weathering and erosion of the bedrock, glacial transport, and settling. Subsequently, ground water and surface water quality are affected by the lithological source.

The Ganga-Brahmaputra-Meghna (GBM) river basin, which have an area of 1.7 million km^2 is drained jointly by the River Ganges, River Brahmaputra (also known as River Jamuna in Bangladesh), River Meghna and their numerous tributaries and distributaries. The GBM basin have a population of more than 150 million and considered as the world largest fluvio-deltaic systems and one of the most populous regions of the planet. In recent few decades, with the increasing demand of ground water for domestic, irrigation (round the year for food production), industry and the growing population rate led the extensive exploitations of fresh and potable ground water. Beside this, indiscriminate use of surface water and the introduction of high-yielding dry-season agricultural activities accelerated the demand for irrigation in the GBM basin (Harvey et al. 2002). This led to the shift of water supply policy from surface water to ground water in these areas. As a consequence, several million wells (ranging from domestic hand pump to motor-driven deep tube-well) were installed within the area to meet drinking, irrigation, and industrial water demands (Smith et al. 2000; BGS/DPHE/MML 2001; Harvey et al. 2005). However, in the present scenario, a large part of the GBM basin ground water was determined to have elevated concentrations of arsenic (As), which can impact human health at concentrations of 10 μg/l in drinking water (WHO 1993). Regarding the source of such high level arsenic, it has been hypothesized that the non-point source, geogenic As, mostly occurs in the Holocene shallow aquifers of the GBM basin and probably has been mobilized from the sediments by redox reactions (e.g., Saha 1991; Bhattacharya et al. 1997; CGWB 1997; Nickson et al. 1998; BGS/DPHE/MML, 2001; McArthur et al. 2001, 2004; Ravenscroft et al. 2001; Harvey et al. 2002; Mukherjee 2006).

In recent years researchers have published numerous report that the drinking water wells have elevated concentration of arsenic in various parts of middle Ganga plain (MGP) mainly eastern part of U.P and Bihar. The present study aims at identification of arsenic safe aquifer along the vulnerable and arsenic affected zones/regions in Eastern U. P. (Ballia district). As proposed, within the time frame the project, detailed hydrogeochemical characterization of the ground water and surface water of Ballia district have been done. It was attempted to identify different highly arsenic affected villages along with the villages which have found to be safe. Furthermore, the results revealed that major portion mainly in

the newer alluvium plain shallow depth ground water of Ballia district are not safe as prescribed by International and National standards, for drinking purposes.

2. Literature review

The Ganges River basin (GRB), the water machine of India houses more than 8.3% of world population, with an area of 1.086 million km^2 and represents various contaminants which creates severe water related challenges in the basin (Amarasinghe et al., 2016). Most of the occupying parts of Gangetic basin (in Uttar Pradesh (UP), Bihar and West Bengal) is extremely fertile for agricultural practices and have a very favorable condition for urbanization and industrialization. The large-scale vulnerability of surface water by industrialisation, urbananization in last few decades (ideally after 1970), pushed community to use ground water as major source water for drinking and irrigation within the basin (Fendrof et al., 2010; Harvey etal., 2002). As a result, despite the positive impacts due to high withdrawal of ground water, significant portion of the GRB basin has been reported with geogenic and anthropogenic contamination. Of these, the pollution that has attracted maximum national and international attention is the elevated concentration of arsenic (As) in ground water of the vast alluvial plains of GRB basin. In Indian subcontinent, the widespread symptoms of disease in people drinking As-contaminated ground water have observed and severe epidemiological studies conducted in these areas the risk of cancers of the liver, bladder, and lung (GuhaMazumder 1998; Fendrof 2010). Drinking of As-containing ground water may also cause cardiovascular disease and inhibits the mental development of children (Fendrof et al., 2010). In the last few decades numerous literature have been published on As contamination in the GRB Basin, (Edmunds et al., 2015; Smedley and Kinniburgh 2002), especially in the India and Bangladesh. In India, GRB have been divided in four parts, i.e., Trans gangetic plain (TGP), upper ganga plain (UGP), middle ganga plain (MGP) and lower ganga plain (LGP) based on its catchment area.

In recent years researchers have published numerous report that the drinking water wells have elevated concentration of arsenic in various parts of MGP and LGP, mainly Ballia, Varanasi, and Gazipur District in UP (e.g., Shah, 2008; Kumar et al., 2009), Bhojpur district in Bihar (Chakraborti et al., 2003), Sahibganj district in Jharkhand (e.g. Bhattacharjee et al., 2005;) and Malda, Murshidabad, Nadia, North-24-Parganas, South-24-Parganas, Howrah, Hooghly and Bardhaman in West Bengal (Sengupta 2009). There are numerous wells containing As >10µg/liter at all depths in the above mentioned districts and extensive spatial variability of As concentrations have been observed at shallow depths, even within a single village (Fendrof et al., 2010). This extensive variability makes researchers difficult to

compare different field site and to identify the common biogeochemical and hydrological process that regulate As levels in ground water (Fendrof et al., 2010).

Source of Arsenic in ground water can be classified as: i) natural or geogenic; and ii) anthropogenic. There are few commonly identified minerals like arsenopyrite, orpiment, realgar, claudetite, arsenolite, pentoxide, scorodite and arsenopalledenite, from which As can originate (Smedley and Kinniburgh 2002). Flood deposits during sedimentary formations also causes As occurrence. However, in most of the literature arsenopyrite/As-rich pyrite is reported as a commonly abundant mineral (Smedley and Kinniburgh 2002; Welch et al. 1988). Beside this anthropogenic sources and human activities also accelerate the release of naturally existing As (BGS 2000; Neidhardt et al. 2013). There have been several significant hydrological and geochemical studies in Bangladesh (BGS/DPHE, 2001; McArthur et al., 2001; Ravenscroft et al., 2001; Harvey et al., 2002; van Geen et al., 2003; Zheng et al., 2004) and fewer in West Bengal (e.g. CGWB, 1997; Chakraborti et al., 2001; McArthur et al., 2004; Mukherjee, 2006) to delineate the source of the contamination and understand the cause in order to find a arsenic free aquifer and suggest the preventive and remedial measures. In MGP major source of As is geogenic and most of the highly As-contaminated wells located on younger alluvium plain (Mallick and Rajagopal 1995). Most of the researcher accepted the hypothesis that that As is migrated with fluvial sediments from Himalaya during erosion and transport of Himalayan sediment further leads to downstream deposition of As. Arsenic release mechanism and transportation within the aquifer is regulated by different factors. The main hypotheses that have been proposed for explaining the source of the contamination include: 1. Oxidation of pyrite, 2. Reductive dissolution of Iron oxyhydroxides; 3. Reduction and reoxidation; and 4. Competitive ion exchange (Bhattacharya et al. 2011; Das et al. 1995; Mandal et al. 1996; Ravenscroft et al. 2009; Rahman et al. 2001; Roy and Saha 2002). There is indeed a positive relation have been observed between As and Fe in the sediment samples of Ganges-Brahmaputra-Meghna basin (Fendrof et al., 2010; Zheng et al., 2004). Destabilizing As on Fe-oxide phases is one of the key geochemical processes which control the contamination of ground water (Zheng et al., 2004). Microbial reduction of Fe oxides releases As into the dissolved phase under anoxic condition (Islam et al., 2004). On the other hand dissolution of Fe-oxides associated with the releases of competitive ion like phosphate during adsorption (BGS/DPHE, 2001). Microbial reduction of Fe(III) and As(V) reduction requires a supply of adequate organic carbon and

depends on the microbial metabolism and biological oxygen demand of the system (Fendrof et al., 2010; Harvey 2002).

The shallow aquifers of LGP have been identified with strong reducing condition, developed as a result of the surficial clay layers (wherever present) which restricts the inflow of oxygen into the aquifer (Smedley and Kinniburgh 2002), which indicates the redox conditions within the aquifers correspond to the As(V) and Fe(III) reduction process. However, liberation of the entire sorbed load of the iron hydroxide (which includes arsenic) into the ground water is restricted in some shallow (<20 m) aquifers where sulfate supplied by recahrged water. Further, the promoted sulphate reduction process by organic carbon produces sulphide, which can bind As and formed soluble sulfides mineral that effectively remove As from ground water.

Arsenic contamination of tube wells in the middle Gangetic plain was first reported in 2002 in Semria Ojha Patti village (area 4 km^2), Sahapur block in the Bhojpur district of Bihar, India (Chakraborti et al., 2003). It is reported that 57 blocks in 15 districts of Bihar are As-affected (Saha et al., 2010). Bhattacharjee et al (2005) reported arsenic contamination in three out of nine blocks surveyed in Sahibgunj district, Jharkhand. Several attempts have been made to investigate the concentration of As in ground water, health effect due to As toxicity and the mechanism of mobilization process of As in the middle Ganga plain of Bihar (Chakraborti et al., 2003, Kumar et al., 2010) Arsenic above 50 µg/L was detected in 47% of tubewells in Bhagalpur, Bihar (Kumar et al., 2010), while it was 58.6% in Semaria Ojha Patti, Bihar, India (Chakraborti et al., 2003).

Most recently, Ahamed et al. (2006) conducted a survey in Ballia, Gazipur and Varanasi districts of Uttar Pradesh. Analyses of 4780 tube well water samples revealed that arsenic concentrations exceeded 10 µg/L in 46.5%, 50 µg/L in 26.7% and 300 µg/L in 10% of the samples. It is reported that around 4.4% of the surveyed sources contained arsenic concentration >50 µg/L in Ballia district (Nickson et al., 2007). Water samples from India Mark II hand pumps, private hand pumps and dug wells from Chain Chhapra, Rajpura Ekauna, Chaube Chhapra, Ramgarh Dhala, Reckni Chhapra, Dalan Chhapra and Sughan Chhapra village have been reported with arsenic concentration more than 50 µg/L. These all villages are located in flood plains and younger alluvium and depth of abstraction structure varies from 13 to 64 mbgl MoDWS, 2011). Moreover, these parts of the middle Gangetic Plains are densely populated areas of India and after Independence dependence on ground

water has increased tremendously, which intensifies the likely health problem from ground water contamination. Thus, there is an immediate need for an inventory which can take account of the amount of arsenic present in the area and number of people actually exposed to it.

3. Study Area

Ballia is the eastern most district of Uttar Pradesh comprising seventeen administrative blocks, covering an area of 2981 sq.km, lies in between 25º33' and 26º11'N latitudes and 83º 38' and 84º 39'E longitudes (Figure 1). The district is bounded on the north by Ghagra River and in the south by Ganga River (Figure 2). The study area has an average rainfall of 983 mm with mean annual temperature of 27ºC, which varies between 5.4^{o}C to 41.5^{o}C (MoDWS, 2011). Ballia is an agricultural dominant district. The district has two sources of irrigation: (a) Dharighat Lift Irrigation canal and (b) tube wells (ground water) with a share of 72.61% for ground water and 27.39% of surface water. The entire area forms an interfluves zone of Ghagra and Ganga River and possesses plain flat topography. The geological formations met within the district are gangetic alluvium consisting of older and younger alluvium (Figure 3). The age of these formations range from upper pleitocene to holocene formations (Ravenscroft et al. 2001).

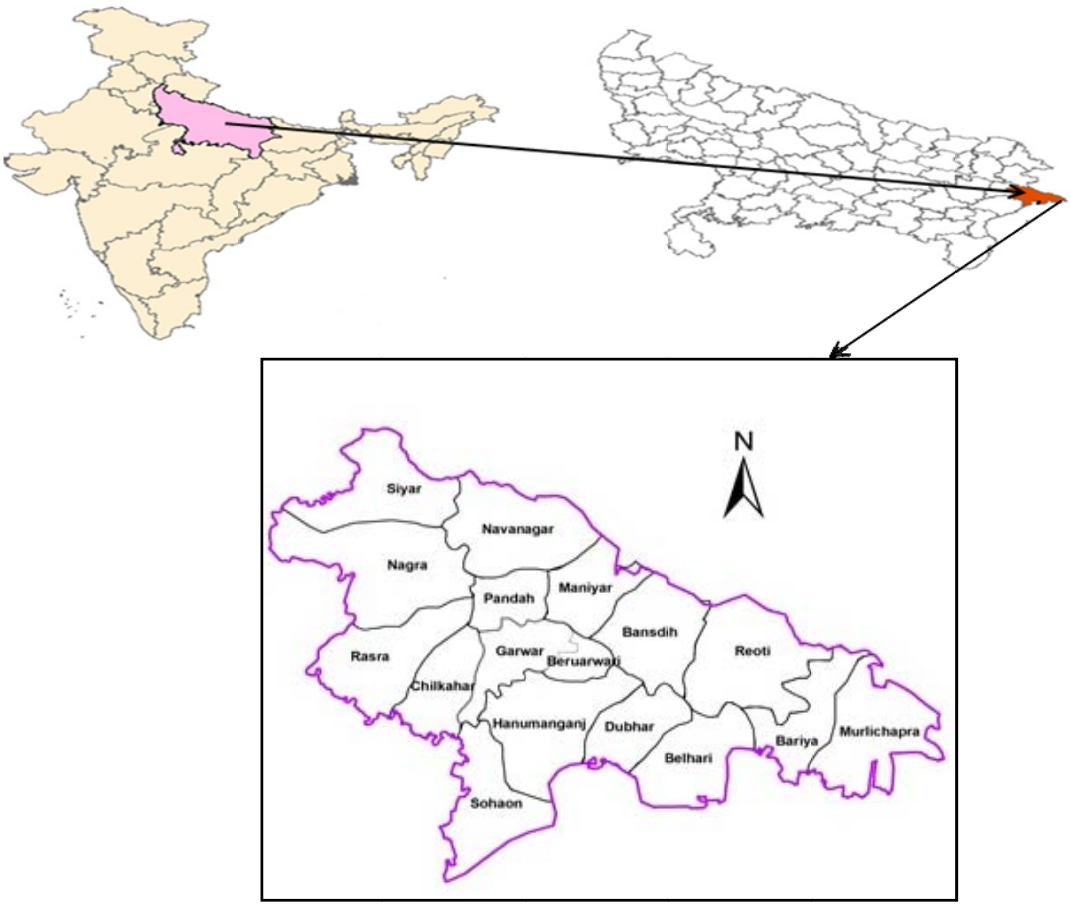

Figure 1: Map showing Ballia district (U.P., India) comprising of seventeen administrative blocks

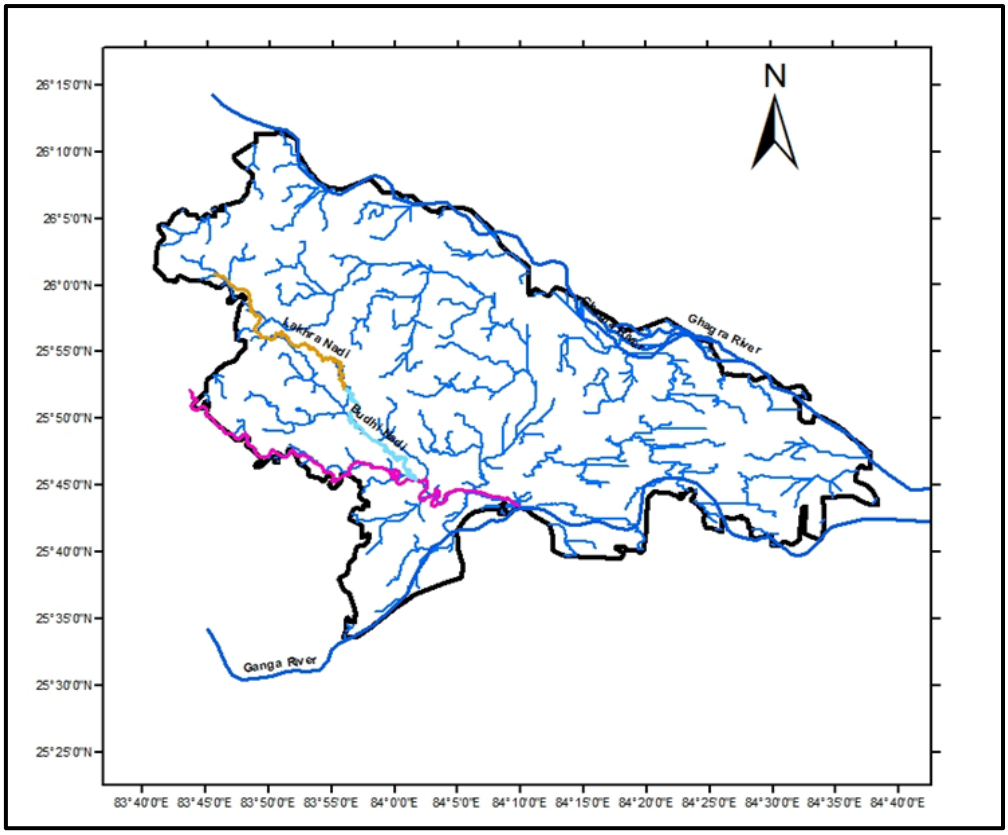

Figure 2: Drainage map of the Ballia district, Uttar Pradesh, India

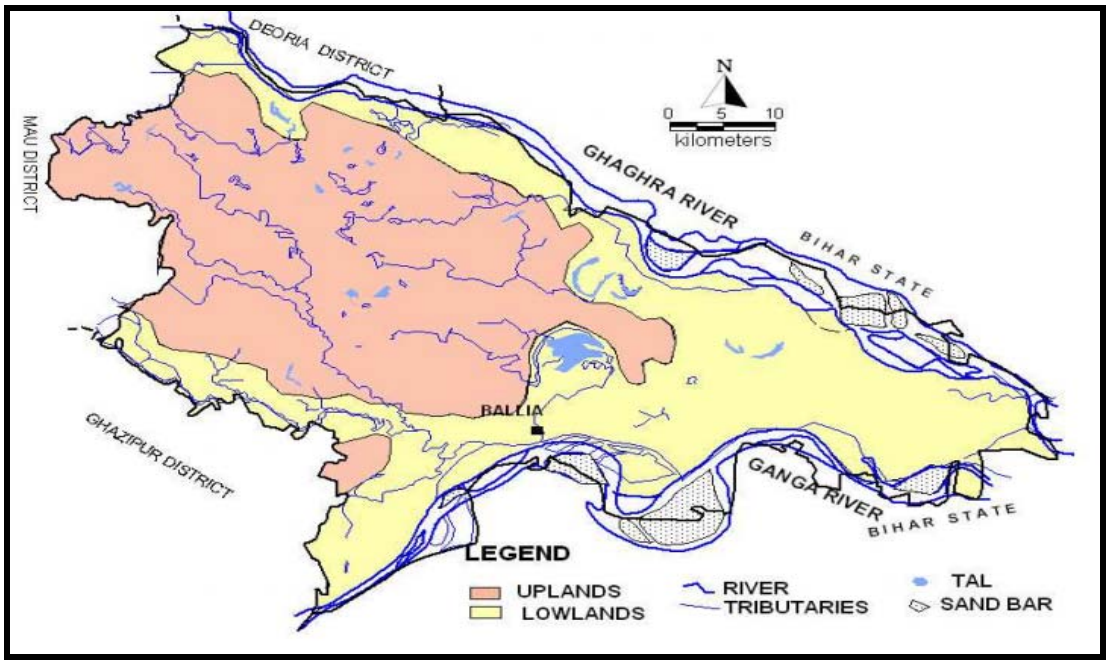

Figure 3: Geomorphology and drainage map of Ballia District (UP) (Source: MoDWS 2011)

The Middle Ganga regions are primarily composed of flood plains and piedmont plains. The holocene alluvial plain of Central Gangetic basins slopes from north to south, interrupted locally by sandy ridges and basins, but looks smoother on a regional scale. The basin has been incised by dendritic drainage and the channels filled by organic muds of recent age deposit (Ravenscroft et al., 2005). The large-scale features of the basin correspond to major climate changes fall in the late Quaternary (Singh 2004). The exploratory drilling by CGWB indicates that these quaternary unconsolidated sediments are underlain by hard rock formations of Archeans age east of Faizabad ridge (MoDWS, 2011). The older alluvium had been really characterized by yellow colour clays, silt rich in calcareous concentrations and nodules, locally known as kankar. It is normally poorly sorted and appears to be less permeable. While, the newer alluvium of the area is found as unconsolidated occurs mainly along flood plains of Ghaghra, Ganga and tons rivers. The alluvium formation is found with poor in calcareous matter, contains lenticular beds of sand gravel and clays throughout the river laid deposits and sandy horizons of the aquifer zones.

4. Methodology

4.1 Construction of conceptual model

The detailed workflows of the study have been presented in the Figure 4. All the workflow components are described in the subsequent paragraphs.

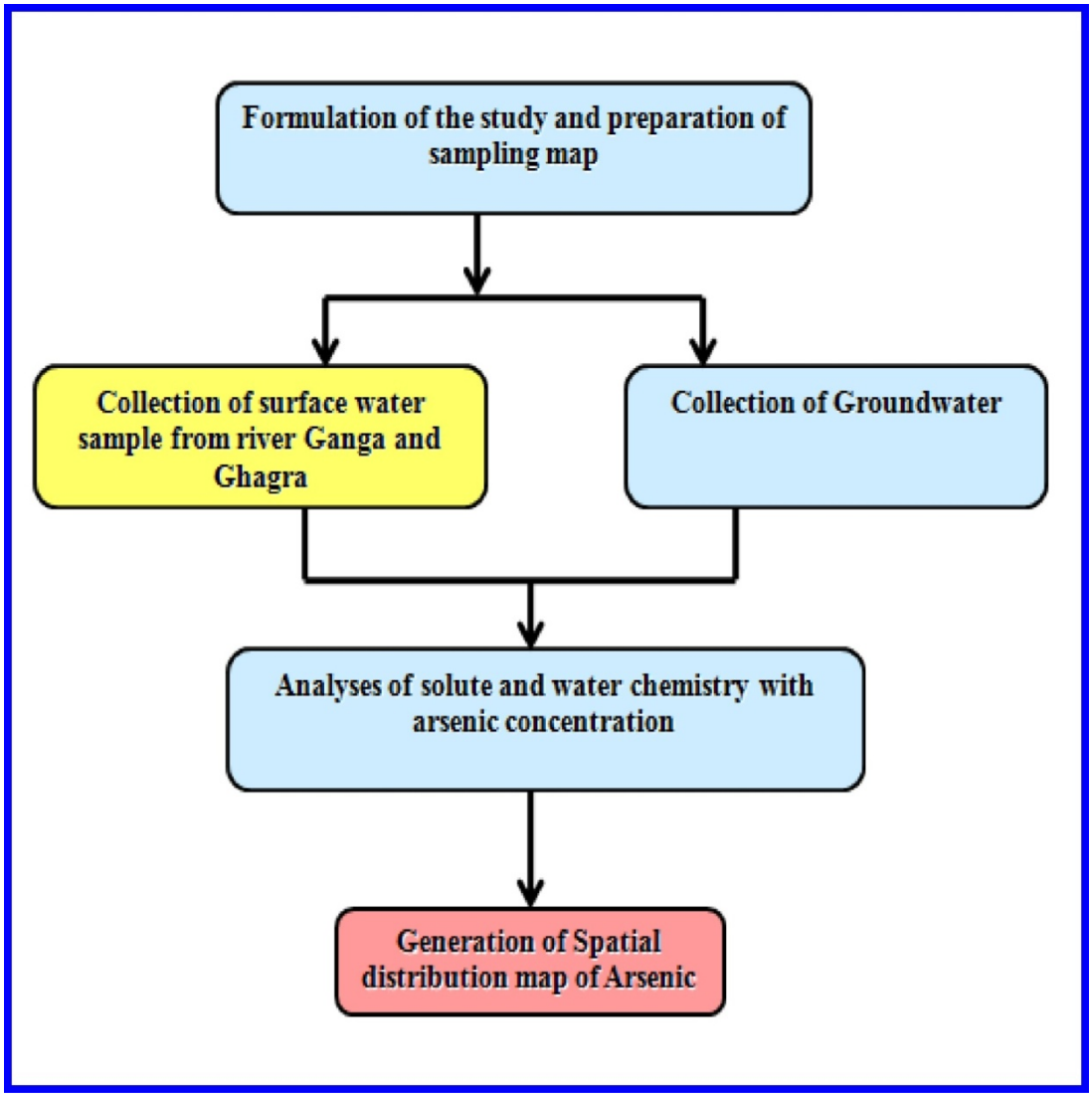

Figure 4: Workflow used for the study

4.2 Ground water and surface water sampling

Ground water and surface water samples were collected in pre-monsoon (May, 2016) season from India-marked-hand pump (110-140 ft depth) for all the seventeen administrative blocks of district by making grid of size 4 km x 4 km (Figure 5). The details of sampling location along with pH, EC and As concentration has been given in Table 1, However, the

interpretation of results have been described in section 5. Some field photographs are shown in Figure 6.

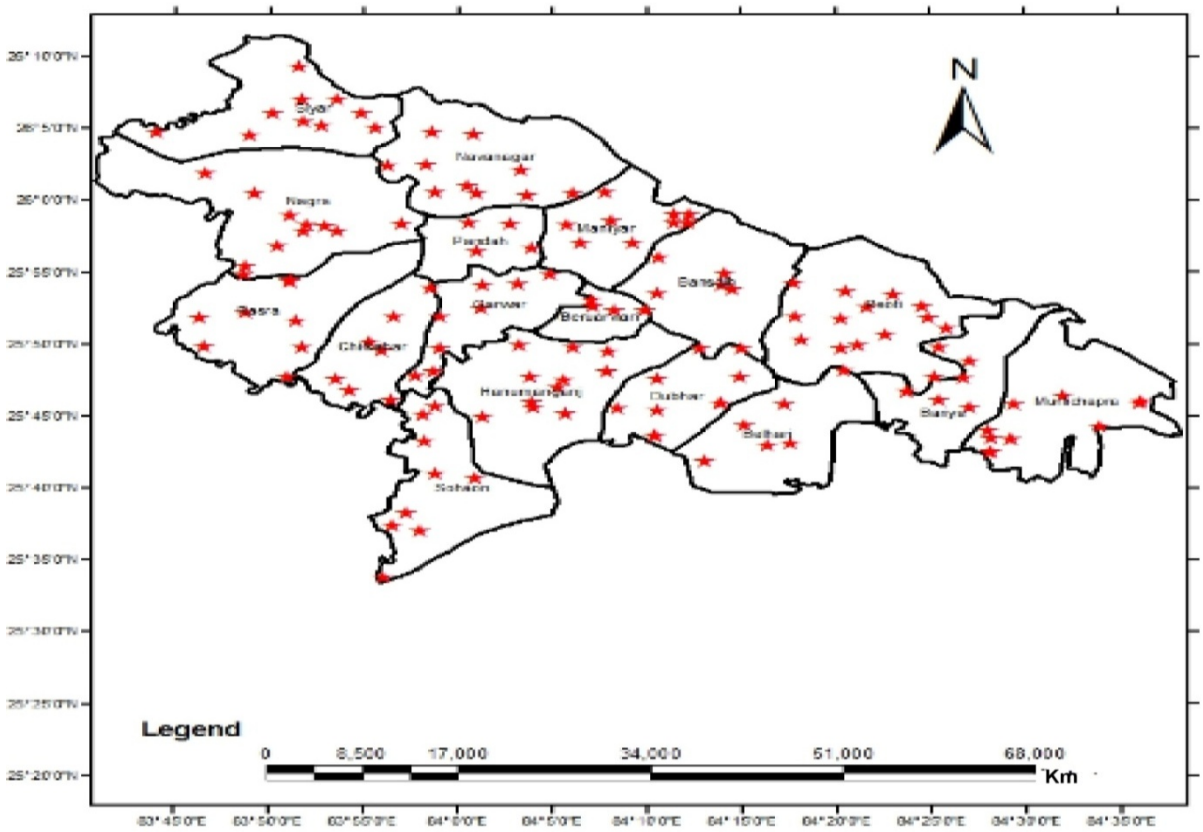

Figure 5: Sampling locations in all the seventeen administrative blocks of Ballia

Table 1: Sampling location details along with pH, EC and As concentration

Sample Id	Villlage name	Block	Lattitude	Longitude	pH	EC (µs/cm)	As (ppb)
1	Muhamedpur	Dubhar	25°43.730	84°10.540	7.23	625	34.44
2	Ganga River (Muhamedpur)	Dubhar	25°43.681	84°10.488	7.88	578	6.72
3	ShivpurDiar	Dubhar	25°41.922	84°13.142	6.73	856	245.99
4	Jamuan	Dubhar	25°45.464	84°10.583	6.54	811	15.64
5	Pipra	Dubhar	25°45.946	84°13.921	7.18	659	29.91
6	Pipra	Dubhar	25°45.996	84°14.035	7.14	681	141.03
7	Vishnupura	Dubhar	25°44.409	84°15.214	7.19	707	46.37
8	Patkhauli	Dubhar	25°47.787	84°15.005	7.19	378	155.96
9	Shankarpura	Dubhar	25°47.694	84°10.586	7.16	726	172.42
10	Umraoganj	Dubhar	25°45.642	84°08.507	7.11	801	154.33
11	Bahuara/ChiranjiChapra village	Murlichapra	25°43.521	84°28.275	7.27	618	164.06
12	Damodarpur	Murlichapra	25°42.543	84°28.273	7.1	865	44.02
13	Ganga River(at Damodarpur)	Murlichapra	25°42.483	84°28.182	8.12	558	3.19
14	Shivpur	Murlichapra	25°43.436	84°29.312	7.19	844	15.64
15	Shivpur	Murlichapra	25°43.440	84°29.286	7.02	1451	25.73
16	Sonbarsa	Murlichapra	25°45.893	84°29.521	7.41	593	168.65
17	Naukatola	Murlichapra	25°46.448	84°32.093	7.57	694	73.97
18	Khakhankatola	Murlichapra	25°44.288	84°34.056	7.97	289	26.37
19	Ghaghra River (Athagawan)	Murlichapra	25°46.068	84°36.197	8.1	313	2.75
20	Athgawn	Murlichapra	25°45.936	84°36.152	7.35	615	77.66
21	HaldirampurChati(Bazaar)	Siyar	26°06.174	83°54.925	7.51	492	5.22
22	BAHUTA (updhyapur)	Siyar	26°05.113	83°55.702	7.29	592	2.25
23	Sonbarsa (Rasmohanpur)	Siyar	26°07.069	83.53.603	7.29	527	13.25
24	Turtipara	Siyar	26°09.349	83°51.557	7.12	647	15.95

25	Ghghra River(near Turtipara)	Siyar	26°08.69	83°52.127	7.97	294	2.43
26	Chaukia Mod	Siyar	26°07.104	83°51.784	7.29	637	7.40
27	Shahpur (Atgara)	Siyar	26°05.231	83°52.840	7.38	516	3.25
28	Pharsatar	Siyar	26°05.532	83°51.840	7.56	537	4.21
29	Khunta	Siyar	26°04.823	83°44.048	7.36	648	6.37
30	Pasuhari	Siyar	26°04.611	83°49.045	7.46	500	8.68
31	Dubhauli	Siyar	26°06.170	83°50.213	7.27	615	6.43
32	Hansnagar	Belhari	25°43.181	84°17.636	9.57	554	42.69
33	Chain Chapra	Belhari	25°43.051	84°16.426	7.33	599	32.51
34	Basudharpat	Belhari	25°45.912	84°17.302	7.26	809	21.26
35	Aghaila	Reoti	25°48.253	84°20.486	7.09	716	651.69
36	Gaighat	Reoti	25°49.990	84°21.200	7.45	426	18.65
37	Reoti	Reoti	25°50.730	84°22.667	7.23	719	117.72
38	AsmanThota	Reoti	25°49.777	84°20.347	7.34	570	181.24
39	Sahtwa (Thana Mod)	Reoti	25°50.401	84°18.209	7.42	476	17.40
40	Gangapur (Ramgarh)	Belhari	25°46.736	84°23.784	7.01	758	29.33
41	Ghagra River(Ramgarh)		25°46.736	84°23.784	7.5	578	1.58
42	Prasad Chapra	Beriya	25°46.184	84°25.486	7.03	747	48.62
43	BhawalChapra (Murlichapra)	Murlichapra	25°44.056	84°28.140	7.07	635	203.24
44	Tengarahi	Beriya	25°45.633	84°27.151	6.96	903	146.31
45	Dalatpur	Beriya	25°47.738	84°26.814	7.26	487	111.29
46	Naukagaon	Reoti	25°47.774	84°25.298	7.34	406	170.76
47	Siyarahiyan Village	Beriya	25°48.887	84°.27.127	7.19	585	229.27
48	Srinagr	Beriya	25°49.878	84°25.543	7.1	690	49.97
49	Barutarpar	Reoti	25°51.176	84°25.942	7.18	616	219.71
50	Rati Chapra	Reoti	25°51.889	84°24.993	7.37	466	94.88
51	Ghagra/Saruya	Reoti	25°52.682	84°24.618	7.73	294	15.79
52	Bhopalpur (Gamsahitola)	Reoti	25°53.536	84°23.098	7.32	439	187.16
53	Maharjpur (Bharula)	Reoti	25°53.710	84°20.617	7.29	512	90.06

54	Harida Kala	Reoti	25°52.671	84°21.761	7.16	588	74.62
55	Naina	Reoti	25°51.851	84°20.377	7.21	604	35.70
56	BariarpurChati	Bansdih	25°51.960	84°17.911	7.27	574	13.86
57	Bilariya	Sohaon	25°40.786	84°0.942	7.56	559	45.02
58	Bahaghuna	Sohaon	25°37.090	83°58.050	7.07	584	159.18
59	Tutuyari	Sohaon	25°38.363	83°57.314	7.14	773	145.02
60	Ekauni (Dularampur)	Sohaon	25°37.449	83°56.613	7.01	1012	451.10
61	Narianpur (Kotwa)	Sohaon	25°33.885	83°56.097	7.01	822	28.94
62	Itahi	Sohaon	25°41.034	83°58.850	7.05	711	122.21
63	Singarpur	Sohaon	25°43.331	83°58.313	7.18	584	132.80
64	BiwipurChati	Sohaon	25°45.138	83°58.224	7.21	479	25.73
65	Nagagawn	Sohaon	25°45.777	83°58.873	7.31	938	26.08
66	Chitwara	Sohaon	25°44.994	84°01.337	7.29	529	80.40
67	Jagdishpur	Beruanwari	25°52.407	84°08.347	7.31	636	11.93
68	Dhaidhara	Beruanwari	25°52.763	84°07.242	7.07	551	66.08
69	AhirKatola *(BaswantKatola)	Beruanwari	25°53.168	84°07.158	7.2	443	36.43
70	Kaithauli	Beruanwari	25°52.434	84°09.966	7.15	465	16.08
71	Narainpur	Beruanwari	25°53.578	84°10.634	6.96	1016	119.30
72	Deorar	Bansdih	25°56.088	84°10.670	7.15	669	36.08
73	SultanpurChati	Bansdih	25°58.530	84°12.276	7.08	714	27.08
74	Kisounipur	Bansdih	25°58.530	84°12.276	6.87	823	16.43
75	ParvatPurChati	Bansdih	25°55.010	84°14.194	7.26	589	123.97
76	Suaraha	Bansdih	25°54.303	84°17.814	7.24	575	128.64
77	Pakharia Tal	Bansdih	25°54.226	84°14.044	7.47	775	13.22
78	Kharauni (Rajgaon)	Bansdih	25°53.932	84°14.611	10.23	653	21.45
79	Amdur	Bansdih	25°49.775	84°15.108	7.33	488	106.08
80	sahodihChoti	Bansdih	25°49.817	84°12.889	7.17	640	42.86
81	Khalilpur	Chilkahar	25°49.667	83°56.014	7.25	737	7.89
82	Sawrain (SawrainChoti)	Chilkahar	25°50.225	83°55.389	7.07	933	1.61

83	Domari (Dumari)	Chilkahar	25°47.912	83°57.782	7.17	627	3.22
84	Nasirpur	Chilkahar	25°46.169	83°56.526	7.33	506	6.43
85	Naferpur	Chilkahar	25°46.868	83°54.300	7.58	713	6.43
86	Dongain(Tilkadauri)	Chilkahar	25°47.680	83°53.615	7.17	804	0.32
87	Dehri	Rasra	25°49.840	83°51.833	7.3	592	1.29
88	Juvania	Rasra	25°47.784	83°50.992	7.51	705	6.43
89	Nasirpur	Rasra	25°52.250	83°48.763	7.14	663	17.61
90	MustafabadChati	Rasra	25°51.884	83°46.379	7.04	964	0.00
91	Dulhapur (Dulhapur Primary school)	Rasra	25°49.940	83°46.652	7.48	416	6.43
92	Shah Mohammadpur (Jakaria)	Rasra	25°54.573	83°51.171	7.07	682	3.22
93	Athilipur (athili)	Rasra	25°54.420	83°51.171	7.22	728	6.43
94	Chhtouni/rasra	Rasra	25°51.674	83°51.460	7.39	1256	3.22
95	Bhadapa	Chilkahar	25°51.957	83°56.697	7.34	433	0.11
96	Chanda Dih	Chilkahar	25°52.566	84°01.302	7.48	547	7.72
97	Bishar	Pandah	25°56.727	84°03.972	7.21	640	6.32
98	Sonadi	Pandah	25°56.532	84°01.062	6.97	692	6.41
99	Paharajpur	Pandah	25°57.073	83°59.668	7.23	527	0.32
100	UshashBawarani	Pandah	25°58.995	83°58.755	7.4	656	3.22
101	Chakra	Pandah	25°58.538	84°00.609	7.04	788	1.66
102	Usraila	Pandah	25°58.440	84°02.783	7.19	649	3.22
103	Khudiatola	Navanagar	26°01.099	84°0.556	7.15	614	0.00
104	Shikandarpur	Navanagar	26°02.226	84°03.412	7.24	481	11.29
105	Balupur	Maniyar	26°00.587	84°06.172	7.25	569	6.43
106	BichhiBhoj	Pandah	26°00.423	84°03.694	7.28	584	13.25
107	narhanChouraya	Navanagar	26°04.703	84°00.836	7.46	450	3.11
108	Harsan	Navanagar	26°04.834	83°58.653	7.38	848	13.56
109	Sakia	Navanagar	26°02.519	83°58.368	7.16	640	0.00
110	DakuliChati/Dukuli	Navanagar	26°02.474	83°56.291	6.99	468	28.94

111	Dakinganj	Pandah	26°00.660	83°58.858	7.33	577	1.61
112	BarwaCharwa	Pandah	26°00.562	84°01.034	7.25	442	18.95
113	Hanumanganj Market	Hanumanganj	25°48.186	84°07.957	7.28	517	16.43
114	Basantpur	Hanumanganj	25°49.609	84°08.014	6.98	614	3.26
115	Subaibandh	Hanumanganj	25°49.841	84°06.142	7.28	499	11.56
116	Kharicha	Hanumanganj	25°50.003	84°03.347	7.01	732	4.50
117	Alwalpur	Hanumanganj	25°47.559	84°05.641	7.15	637	3.22
118	Bhikampur	Garwar	25°47.071	84°05.379	7.18	531	0.00
119	Bisukia	Garwar	25°47.777	84°03.867	7.41	587	4.50
120	Kapuri/Kapuri-Naryanpur	Hanumanganj	25°46.068	84°03.953	7.24	593	3.22
121	Beduli	Hanumanganj	25°45.684	84°04.074	7.22	618	7.89
122	Sagarpali	Hanumanganj	25°45.252	84°05.786	7.32	1047	6.43
123	Patpohar/patopor	Garwar	25°54.907	84°04.905	7.16	634	0.96
124	Ratsar	Garwar	25°54.244	84°03.243	7.2	631	3.22
125	Amderia	Garwar	25°54.190	84°01.344	7.43	1528	11.25
126	Sawan	Chilkahar	25°53.944	83°58.571	7.5	535	8.04
127	BalseraChati	Chilkahar	25°51.978	83°59.079	7.24	631	1.61
128	Hariharpur	Chilkahar	25°49.816	83°59.079	7.65	898	0.64
129	Aundi/audi	Chilkahar	25°48.141	83°58.805	7.05	681	22.54
130	Sarayachawt (Saraichaut)	Nagra	25°57.929	83°51.906	7.24	654	16.43
131	Sonapali	Nagra	25°56.929	83°50.458	7.09	718	3.22
132	Atrauli	Nagra	25°55.492	83°48.796	7.33	659	6.11
133	Shah Mohammadpur	Rasra	25°54.919	83°48.710	7.53	644	3.22
134	Bhimpura/ BarewaBhimpura	Nagra	26°01.970	83°46.623	7.33	582	1.61
135	Madari	Nagra	26°00.541	83°49.302	7.27	599	63.51
136	Parshurampur	Nagra	25°59.041	83°51.145	7.28	528	8.04
137	Chachaiya	Nagra	25°58.381	83°52.018	7.46	681	6.43
138	Bhitkhuna Mode	Nagra	25°58.268	83°53.030	7.45	412	11.29
139	Jajlapur	Nagra	25°57.952	83°53.613	7.49	602	11.23

140	Chitupali	Nagra	25°58.441	83°57.037	7.66	381	22.51
141	Hataunch	Maniyar	25°58.350	84°05.812	7.23	566	0.64
142	Bhagipur	Maniyar	25°58.681	84°08.148	6.97	677	0.00
143	Ahiraulli	Maniyar	25°57.111	84°06.546	7.26	597	14.45
144	Rampur	Maniyar	25°57.145	84°09.332	7.17	626	6.43
145	Manikpur	Maniyar	25°58.513	84°11.478	7.19	616	3.22
146	Gandauli	Maniyar	25°59.114	84°11.531	7.08	642	10.65
147	Ghagra river(Kukraghat)	Maniyar	25°59.099	84°12.301	7.66	279	1.07
148	Bahadura	Maniyar	26°00.649	84°07.874	7.55	999	12.32

Figure 6: Some photographs during sampling collection

The samples were acquired for major solute analysis after filtering through a 0.45 µm Nylon-66 membrane filter paper and collected in HDPE bottles after a thorough flushing (at least five times). The pH and EC were measured in field using instruments (Hach, HQ30d portable meter). Filtered samples were collected for cation, anion and bicarbonate analysis in different HDPE bottles. Each cation sample bottle was acidified with the 8M nitric acid, which leads to the immediate acidification of the filtered sample to pH~2. Anion samples were collected as both unpreserved and preserved.The unpreserved anion sample was used for bicarbonate analysis and the preserved anion sample for major anion analysis by adding chloroform.

5. Results and Discussion

5.1 Major solutes and physical parameters

The results of ground water and surface water chemical analyses are presented in Table 2. Most of the analysed sample showed a negative imbalance charge of<-10%. pH of the samples ranging from (6.54-8.56, median 7.25) indicating most of the ground water is neutral. Calcium (Ca^{2+}) is found to be dominant cation in ground water samples followed by Na^{+}, Mg^{2+}, and K^{+}.The concentration of Ca^{2+} is found to be in the range of (21-142 mg/L, median 70 mg/L). The concentration of Na^{+} and Mg^{2+} are varied in the range of (8.6-162 mg/L, median 31 mg/L) and (0.7-113 mg/L, median 34 mg/L) respectively. $HCO_3^{-}$is found to be major anion followed by Cl^{-} and $SO4^{--}$.Bicarbonate (HCO_3^{-}) represents the primary source of alkalinity. In the studied samples bicarbonate concentration ranged between 151 to 591 mg/L with the median of 311 mg/L. High HCO_3^{-} concentration in the studied ground water samples are caused by the presence of carbonaceous sandstones in the aquifers and weathering of carbonate minerals related to the flushing of CO_2^{-} rich water from unsaturated zone, where it is formed by decomposition of organic matter. In the samples Cl^{-} concentration varied from 16.4 to 167 mg/L. The high concentration of Cl^{-} in few ground water samples may be related to the infiltration of sewage effluents from the rural households. This also implies the mixing of ground waters from different aquifers with variable Cl− concentrations or its change in evaporation rate during recharge (Hasan et al. 2007). However, in the study area, the high concentration of Cl^{-} suggests interaction of fresh ground water with high dissolve ion contained sediment. The SO_4^{2-} concentration of the ground water samples varied from 6.2 to 121.1 mg/L, where high SO_4^{2-} concentration observed in few ground water samples. This enrichment in few ground water samples suggest that there is a possibility of the breakdown of organic matter and agriculture runoff carrying unutilized SO_4^{2-} (Anderson 1979).

Table 2: Statistical summary of selected parameters and solutes compositions of the ground water for the study area

Solutes	Unit	Minimum	Maximum	Median	IS(10500-2012) Acceptable limit
pH	-	6.54	8.56	7.25	6.5-8.5
EC	µS/cm	279	1528	617	-
TDS	ppm	186	1023	413	500
$HCO3^-$	ppm	154.28	591.	311	-
F^-	ppm	BDL	9.45	0.2	1.0
Cl^-	ppm	16.45	167	25	250
Br^-	ppm	BDL	102	7.8	-
NO_3-	ppm	0.15	22.23	1.1	45
SO_4--	ppm	6.3	121	18	200
Na^+	ppm	8.67	162	31	-
Mg^{2+}	ppm	0.71	113	34	30
K^+	ppm	0.67	32	5	-
Ca^{2+}	ppm	21	142	70	75
Fe	ppm	0.27	12.54	2.03	0.3
Mn	ppm	0	0.72	0.11	0.1
As	ppb	BDL	651	27	10

The scatter diagram between (Ca^{2+}+Mg^{2+}) vs Tz^+ for ground water (Figure 7) showing a strongly positive correlation ((R^2=0.73), and reflects the high abundance of Ca^{2+}+Mg^{2+}) in the ground water. There is a spatial variability in the distribution of all the ions and the variations are significant to the geomorphology and lithology of the study area.

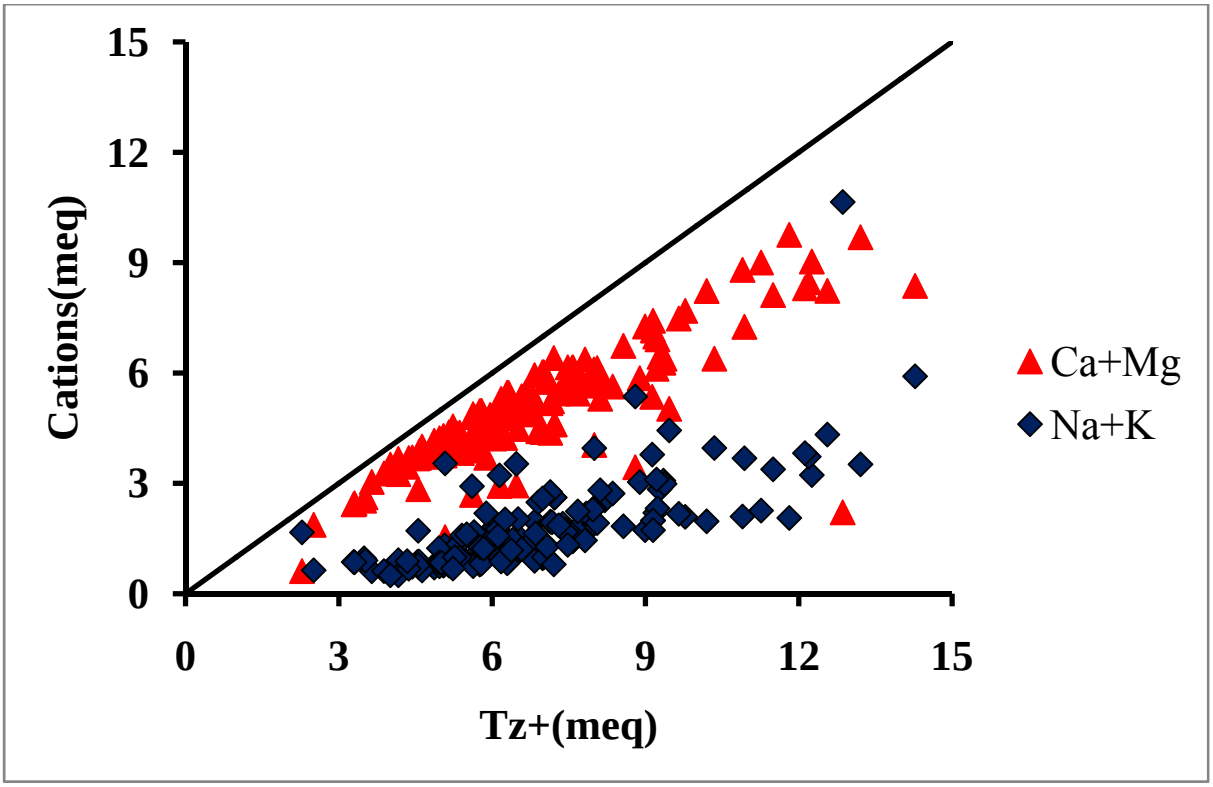

Figure 7: Bivariate plots of Ca^{2+}+Mg^{2+} and Na^{+} + K^{+} Vs total cation (T_{z+}) concentration in the ground water samples. Blacked line is indicating (1:1) equilibrium line for the measured solute.

5.2 Chemical evolution of ground water

The studied ground water from older and younger alluvium plain shows a distinct relationship with major solutes in the ground water. The Piper diagram (Figure 8) suggests that hydrogeochemical facies is of Ca–Mg-HCO3 type Previous published articles by the researchers (Zheng et al., 2004 and Mukherjee and Fryar, 2008; Mukherjee et al., 2012) have already mentioned that in the Middle Gangetic plain high HCO_3^- concentration play important roles in hydrochemical evolution and trace metal mobilization. In the present study we have also observed high HCO_3^- in and around the younger alluvial aquifers.

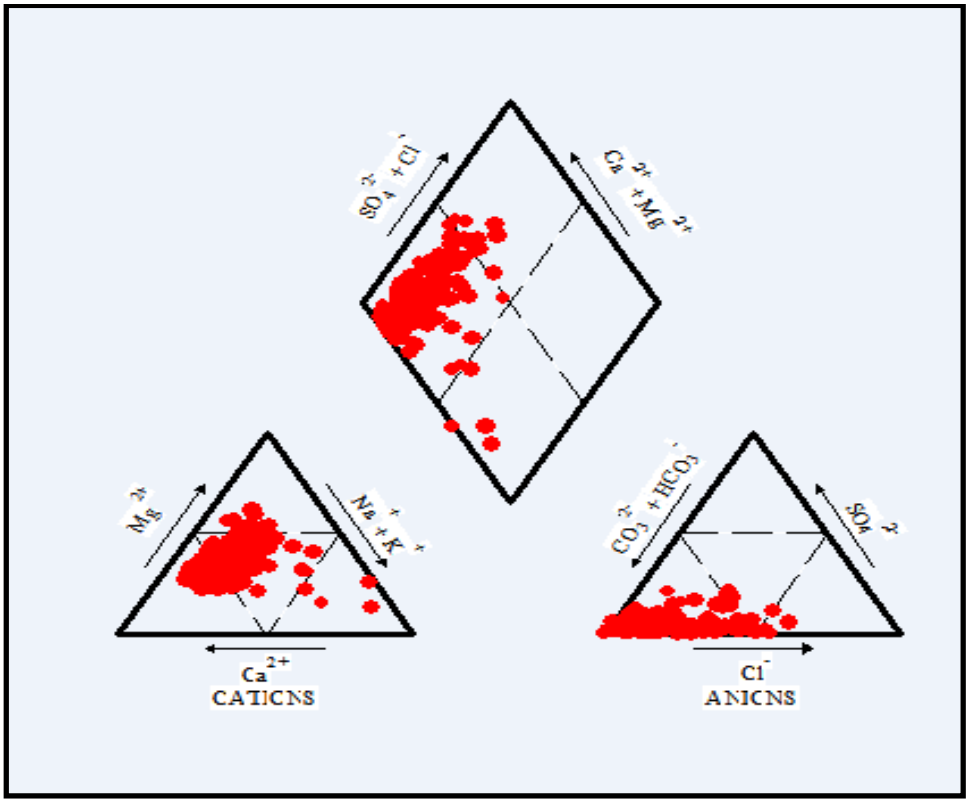

Figure 8: Piper diagram of ground water samples collected from the study area

Hydrogeochemical evolution of ground water depends on, water–aquifer matrix interaction, ground water residence time within the aquifer and the associated chemical process during recharge (e.g. cation exchange (Drever, 1997). Generally, three hydrogeochemical processes control the hydrogeochemical evolution of the ground water when the major solutes intruded to the aquifer matrix: evaporite dissolution, carbonate dissolution, and silicate weathering (Garrels and MacKenzie, 1971; Mukherjee et al., 2010 and Verma et al., 2015). To identify the sources of the major ions in the ground water samples of the study area we have used molar ratios of major solutes for preliminary investigation.. The average $(Ca^{2+}+Mg^{2+})/Tz+$ is 0.75 reflects the abundance of $(Ca^{2+}+Mg^{2+})$ in the ground water and also attributed to carbonate weathering, which is consistent with the carbonate lithology of the area. In Ballia ground water, the average $(Na^{+}+K^{+})/Tz^{+}$ is found to be are 0.15 and infers that the contribution of cations via alumino-silicate weathering is low in comparison to carbonate weathering. The molar Mg^{2+}/Ca^{2+} ratio in study area is observed as ~0.6, infers that there is a less chance of carbonate precipitation.

5.3 Distribution of arsenic

Approximately ~58% of ground water samples collected from the study area are enriched with dissolved As concentrations >10 µg/L (10 µg/L is the acceptable limit for drinking water; BIS, 2012), and it ranges from below detectable limit (BDL) to 651 µg/L (median 27 µg/L). Ground water with elevated As (>100 µg/L) concentration is extensively present in the holocene aquifer of Shivpur Diar (245 µg/L), Pipra (141 µg/L), Umraoganj (172 µg/L), Shivpur (168 µg/L), AsmanThola (181 µg/L), Siyarahiyan Village (229 µg/L), BhawalChapra (203 µg/L), Srinagar (219 µg/L) and Bhopalpur (187 µg/L) areas of Ballia district. In the present study, all the collected ground water samples are of shallow depths (India Mark Hand pump-110 to 140 ft). Hence no variation in vertical distribution of As was observed.

5.4 Relationship of As with other solutes

The ground water samples collected from the study area showed a distinct relationship of As with several other solutes. To calculate the relation of As with other hydrochemical parameters, nonparametric correlation analyses of Pearson correlation was calculated (Table 3).The relation of As with various solutes have been demonstrated in Figure 9, Figure 10, Figure 11 and Figure 12.. Numerous previous studies in lower gangetic plain have described the relationships of As with other solutes (e.g. Bhattacharya et al., 1997, Ravenscroft et al., 2001, Zheng et al., 2004 and Biswas et al., 2011) and combined effects of HCO_3^- and pH in As mobilization from surface of Fe–Mn(OOH) and aquifer sediments (Anawar et al., 2003).. Lot of hypothesis have been deals with the relationship between Fe and As, where many studies showed very close positive co-relationship (e.g. Bhattacharya et al., 1997, Dowling et al., 2002 and Stüben et al., 2003, etc.). Many studies noted that correlations between As and other redox-sensitive parameters (e.g. Fe, Mn, SO_4^{2-}, NO_3) may be limited by the occurrence of multiple reactions in heterogeneous sediments (e.g. van Geen et al., 2004 and Mukherjee and Fryar, 2008). In the present study, all of the ground water samples show very weak correlation of As with pH (-0.165,) (Table 3), which suggest that a redox-dependent mobilization may play important role in As liberation. In the study area, Fe is positively correlated with As, (*0.612)*, but does not show any strong positive correlation with Mn (*0.33,*). The ground water of study area show a moderate positive correlation between As and SO_4^{2-} (0.572), which reaffirms that pyrite/sulphide oxidation may be one of the governing processes for As liberation/mobilization. Accordingly, the calculation of nonparametric correlation suggested that multiple hydrogeochemical processes controls As fate in the

present study area, and no single process can be identified to be predominating the system. High concentration of As is associated with low concentration of HCO_3^- in the studied well. The low concentration of HCO_3^- corresponds to the most important anion species, which competes with As for adsorption sites at mineral surfaces (e.g., Fe/ Mn oxy-hydroxides, clay minerals, and weathered mica), consequently releasing As into the ground water (Ravenscroft 2001).

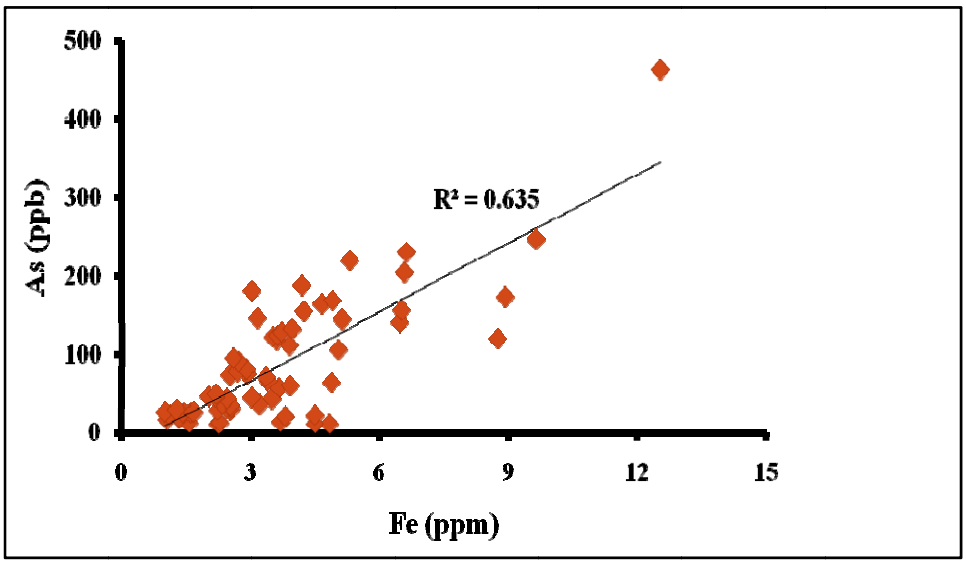

Figure 9: Bivariate scatter plot showing relationship of As (ppb) to Fe (ppm)

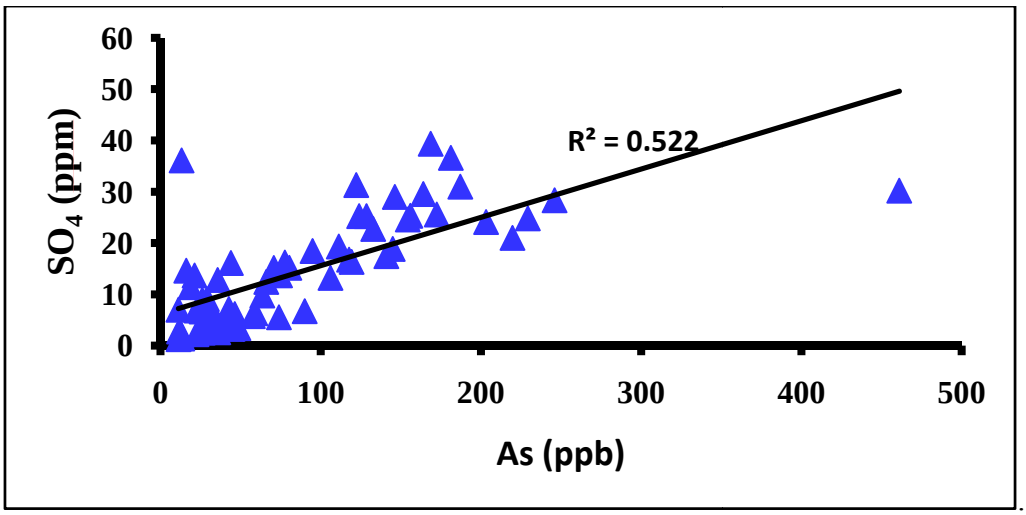

Figure 10: Bivariate scatter plot showing relationship of As (ppb) to SO_4^{2-}(ppm)

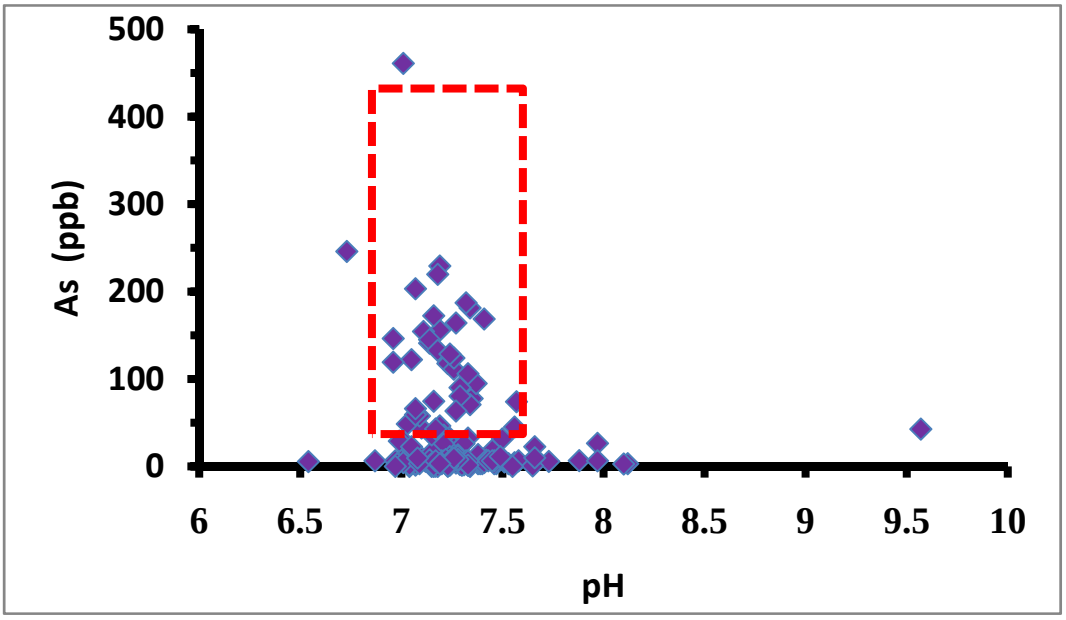

Figure 11: Bivariate scatter plot showing relationship of As (ppb) to pH. Red dashed line indicates the zone of elevated As.

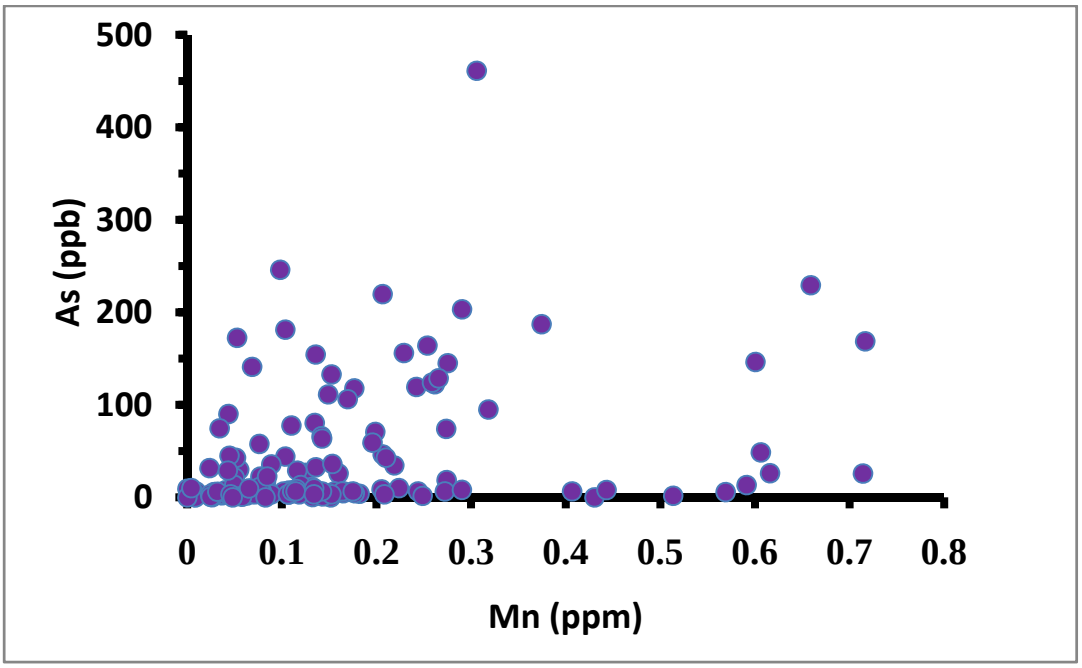

Figure 12: Bivariate scatter plot showing relationship of As (ppb) to Mn (ppm).

Table 3: Pearson's correlation matrix of Ballia ground water samples

Parameter	HCO3-	pH	Na	Mg	K	Ca	Mn	Fe	As
HCO3	1	0.13	0.36	0.364	-.010	.0.396	.015	0.195	0.240
pH	0.136	1	0.11	0.070	0.147	0.038	-0.25	-0.198	-0.165
Na	0.366	0.11	1	0.386	0.093	0.098	0.163	0.080	-0.079
Mg	0.364	0.07	0.38	1	0.18	0.224	-0.191	-0.047	-0.240
K	-0.010	0.14	0.09	0.18	1	0.122	0.062	-0.042	-0.105
Ca	0.396	0.03	0.09	0.24	0.122	1	0.314	0.423	0.288
Mn	0.015	0.25	0.16	0.19*	0.062	0.314	1	0.212	0.333
Fe	0.195	0.19	0.08	-0.04	-0.04	0.423	0.212	1	0.612
As	0.24	0.16	0.07	-0.24	-0.105	0.288	0.333	0.612	1

5.5 Identification of different hot spot of Arsenic

Total As concentrations is extensively present in the ground water of Ballia, typical like a "Hotspot" with a maximum of 651 µg/L in the well of Ekauni (Dularampur) village of Sohaon administrative block of Ballia disctrict, which is close to the Ganges River. The spatial distribution map of Arsenic (Figure 13) shows the location of three hotspot in the Ballia district. These regions are located along the Ganga flood plains (typical hotspot near to convergences of Ghagra and Ganges river), generally in the south and northern east part of Ballia district. These observations suggest that the high As concentrations in the new alluviam plain of the Ganges River are a result of multiple source areas and the likelihood of related mechanisms of mobilization across the entire Middle Gangetic Plain.

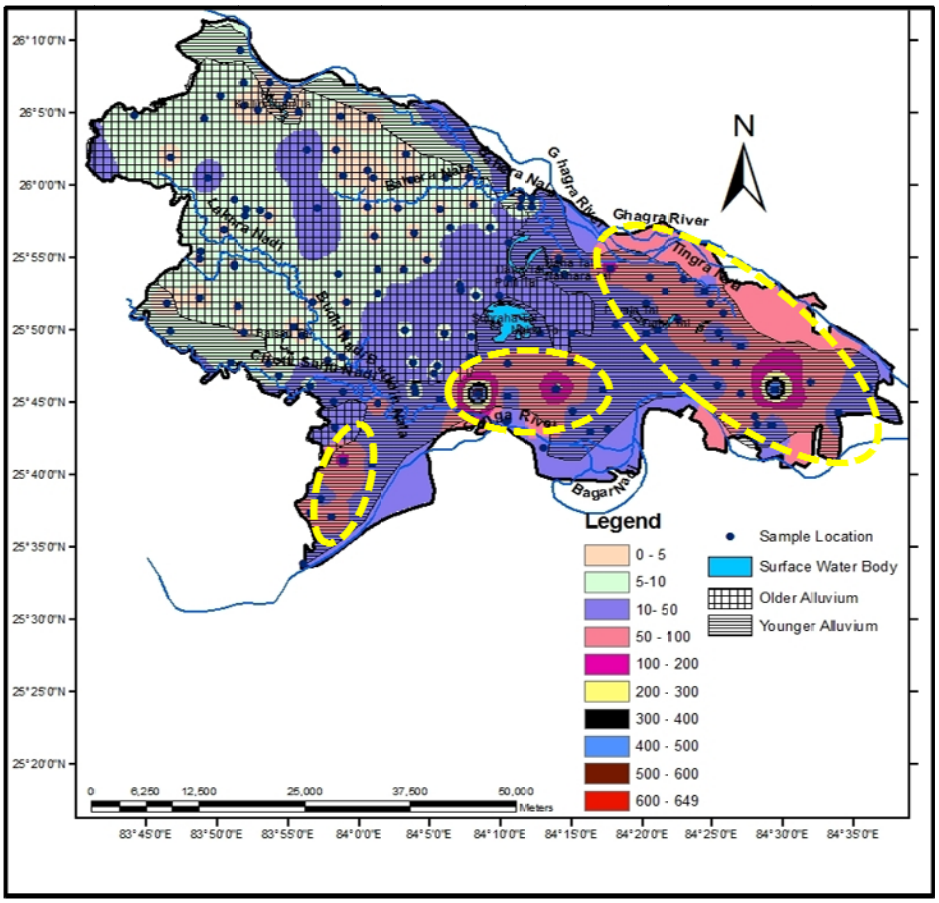

Figure 13 : Spatial distribution of the Arsenic concentration within the Ballia district. The values are provided in µg/L. Yellow coloured dashed circles are indicating the location of the high arsenic contaminated zone.

High arsenic (As) at north-east part of the of the Ballia district could be possibly due to an accumulation of coarser sediment along a Holocene course of River Ganges and River Ghagra or due to some local effects. This indicates that depositional environment and geological age are one of the important factors in controlling As mobilization. Consistently lower As concentrations in older alluvium plain of the Ballia district, which have been identified as safe zone for drinking water supply. This observation may help for better drinking management policy and other water supply and investigation department can target the safe zone in near future. However, a detailed hydrogeochemical and arsenic genesis study is required for better understanding.

6. Conclusions

- The dominant cation was Ca^{2+} followed by Na^{+}, Mg^{2+} and K^{+} and HCO_3^{-} is found to be dominant anion followed by Cl^{-} and $SO4^{--}$. High HCO_3^{-} concentration in the studied ground water samples are caused by the presence of carbonaceous sandstones in the aquifers and weathering of carbonate minerals related to the flushing of CO_2^{-} rich water from unsaturated zone, where it is formed by decomposition of organic matter.

- The strongly positive correlation ((R^2=0.73) between (Ca^{2+}+Mg^{2+}) vs Tz^{+} reflects the high abundance of Ca^{2+}+Mg^{2+}) in the ground water. This is attributed to carbonate weathering, which is consistent with the lithology of the study area.

- In the measured ground water sample, Fe is found to be positively correlated with As (R^2=0.633), likewise, SO_4^{2-} also showed a moderate positive correlation (0.572) between As and SO_4^{2-}, which reaffirms that pyrite/sulphide oxidation may be one of the governing processes for As liberation/mobilisation.

- Arsenic(As) concentration in the studied samples were found in the range of 0- 651 µg/L. Twelve blocks were affected by As contamination (As> 50 µg/L). The enrichment was encountered in the sampling sites that were close to the convergences of Ghagra and Ganges river.

- Approximately ~58% of groundwater samples have As concentration more than acceptable limit of drinking water. Ground water with elevated (As>50 µg/L) concentration is extensively present in the Holocene alluvial aquifers of Ballia (UP) District.

- Consistently lower As concentrations in older alluvium plain of the Ballia district have been identified as safe zone for drinking water supply.

- For better drinking management policy, government and other water supply and investigation department may target the safe zone in near future. However, a detailed hydrogeochemical and arsenic genesis study is required for better understanding.

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