

**TARGETING SAFE AQUIFERS FOR DRINKING WATER  
WITH SPECIAL REFERENCE TO ARSENIC CONTAMINATION IN  
BHAGALPUR (BIHAR) AND GHAZIPUR (UTTAR PRADESH), INDIA.**

*Dissertation submitted to Jawaharlal Nehru University in partial fulfillment of  
the requirements for the award of the degree of*

**MASTER OF PHILOSOPHY**

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2008**



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**CERTIFICATE**

This is to certify that the research work embodied in this dissertation entitled **“Targeting safe Aquifers for drinking water with special reference to Arsenic contamination in Bhagalpur (Bihar) and Ghazipur (Uttar Pradesh), India.”** has been carried out in School of Environmental Sciences, Jawaharlal Nehru University for the partial fulfillment of the award of degree of MASTER OF PHILOSOPHY. This work is original and has not been submitted in part or full for any degree or diploma in any university.

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## *Acknowledgment*

*I wish to take this opportunity to express my deep sense of gratitude to my supervisor, Dr. A.L. Ramanathan, (School of Environmental Sciences, J.N.U., New Delhi) for his untiring supervision, valuable suggestions and inspiring guidance, which enabled me to bring this work into the present shape.*

*I am thankful to Prof. V. K. Jain, Dean of School of Environmental Sciences, J.N.U. for his multidirectional support and academic suggestions. I am also thankful to CIF Staff who helped me to access Centre's Instrument Facility to complete my research work.*

*I would like to give my thanks to ICMR (Indian Council of Medical Research) and KTH(SIDA, Sweden) for giving me financial assistance to carry my research work.*

*I express my special regard to Parijat sir who continuously helped me with brotherhood feelings and even provided his much needed support from dawn to dusk in the crucial hours in analytical part in JNU.*

*I would also like to give special thanks to Gurmeet sir, Rajesh sir, G. Senthil kumar & Jayjit who really gave me technical support (to run various related softwares) for my dissertation work.*

*I also express my gratitude to my seniors Sachin sir, Chander sir, Rita mam, Shruti mam, Sutapa mam, Laxmi, Shashi mam, Bhavna mam & Linda sir who made me to explore various dimensions regarding my dissertation. I am also thankful to Murali for the timely help I received during samples analysis.*

*I am very thankful to my friends Naren, Satish, Ram Avtar, Naseer, Abhijit & Raj kumar, who provided me with important feedbacks, and helped me in encapsulating some tricky aspects related with this work.*

*I also express my hearty feelings to my juniors especially Alok, Mukesh, Ravi & Ram Pravesh for their chirpy faces and morale boosting attitudes.*

*No word can adequately express my indebtedness to my family without whose support and motivation. I would not have reached at this stage of academic distinction.*

*At last my special thanks to almighty BHAGWAN JI.*

*Thank you for all.....*

*Pankaj Kumar*

*To,  
Bhaiya - Bhabhi*

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**G**roundwater : All water which is below the surface of the ground in the saturation zone (below the water table) and in direct contact with the ground or subsoil.

### Why should we study groundwater?

As per fig.1 fresh water accounts for about 2.5% of the total water in the world. Of this 2.5%, less than 0.3% is actually on the surface in lakes and rivers. A much larger percentage of potentially available fresh water is stored in groundwater. Most of this groundwater is neither accessible nor cost effective to recover. The bulk of fresh water (68.6%) is locked in the polar ice caps and also not generally available for use. This means that although there is plenty of water on our planet, only a tiny fraction is potentially available for human use. This enforces us to rethink about this little amount of precious resource.

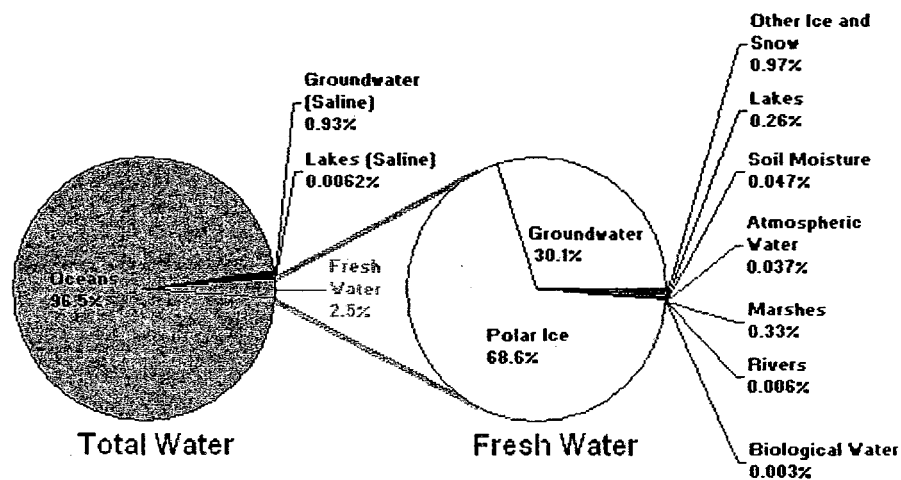


Fig.1. - Global Water Distribution Pattern (UNESCO,1978)

Also groundwater is also an integral part of hydrological cycle (fig.2) as it can be a long-term 'reservoir' of the natural water cycle (with residence times from days to millennia), as opposed to short-term water reservoirs like the atmosphere and fresh surface water (which have residence times from minutes to years). The figure shows how deep groundwater (which is quite distant from the surface recharge) can take a very long time to complete its natural cycle. Groundwater is naturally replenished by surface water from precipitation, streams, and rivers when this recharge reaches the water table, so any disturbance in terms of quality or quantity can affect the whole hydrological cycle.

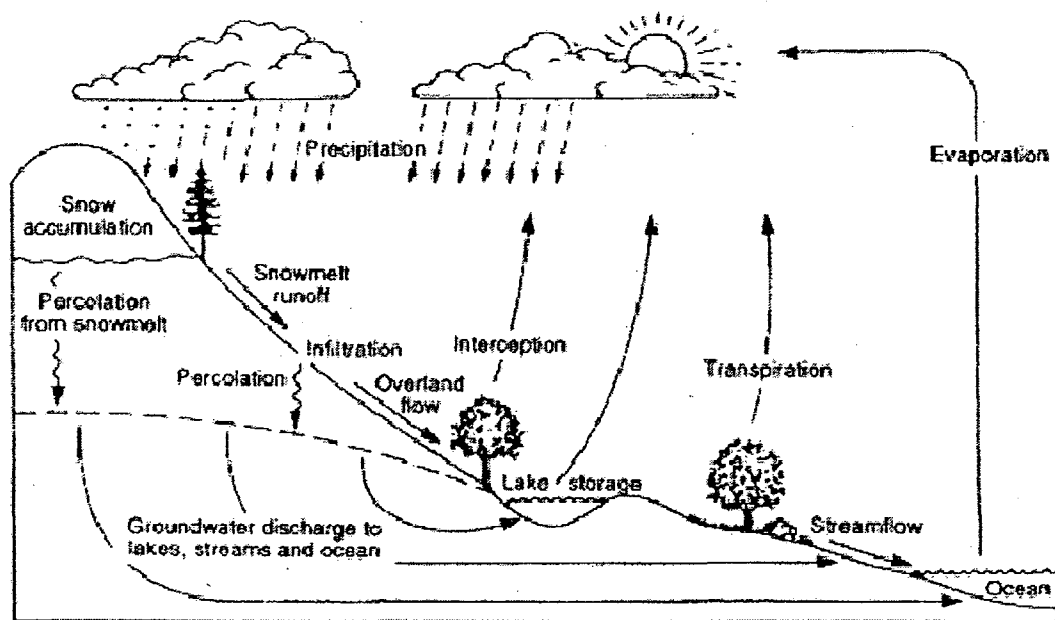


Fig. 2- Hydrological Cycle (USGS)

([www.connectedwater.gov.au/.../hydrological.html](http://www.connectedwater.gov.au/.../hydrological.html))

### Where does the groundwater comes from?

The vast majority of groundwater originates as rainfall. However, not all rainfall ends up as groundwater. Some runs off directly into surface water, some evaporates and the rest soaks into the ground, but has to pass through the soil and underlying strata before it reaches the water table. The ground above the water table is called the *Unsaturated zone*. In this zone some water can be held in storage around soil particles, some flows into drains and into surface water, and some is taken up by plants. The remaining infiltration, known as *Recharge*, eventually reaches the water table and becomes groundwater. There can be a considerable time lag between the fall of rain and recharge to groundwater.

### Where is groundwater found?

Most groundwater is found within aquifers, where it can be exploited via wells or boreholes or it can support other ecosystems such as rivers and wetlands. Groundwater can also be found in unproductive strata but here the groundwater flow is not significant in terms of water supply or the support of ecosystems.

**Aquifer** – A subsurface layer or layers of rock or other geological strata of sufficient porosity and permeability to allow either a significant flow of groundwater or the abstraction of significant quantities of groundwater.

The word aquifer literally means 'water bearer' and refers to a layer of rock or sediment that contains enough accessible water. Water in an aquifer is stored between

the grains of rock. Aquifers can be either consolidated rock (such as sandstone) or unconsolidated (such as the sands and gravels) that make up the high plains aquifer.

### Aquifer types:

#### Unconfined Aquifers

Where groundwater is in direct contact with the atmosphere through the open pore spaces of the overlying soil or rock, then the aquifer is said to be unconfined. The upper groundwater surface in an unconfined aquifer is called the water table. The depth to the water table varies according to factors such as the topography, geology, season and tidal effects, and the quantities of water being pumped from the aquifer. Unconfined aquifers are usually recharged by rain or streamwater infiltrating directly through the overlying soil. Typical examples of unconfined aquifers include many areas of coastal sands and alluvial deposits in river valleys.

Some deeper water bearing units aquifer contain consolidated (e.g., sandstone) layers, and may be separated from the surface by confining layers impermeable enough so that the deep water can be under pressure. Breaches in the confining layer may result in a spring or artesian well flowing at the surface.

#### Confined Aquifers

Confined aquifers are permeable rock units that are usually deeper under the ground than unconfined aquifers. They are overlain by relatively impermeable rock or clay that limits groundwater movement into, or out of, the confined aquifer. Groundwater in a confined aquifer is under pressure and will rise up inside a borehole drilled into the aquifer. The level to which the water rises is called the potentiometric surface. An artesian flow is here water flows out of the borehole under natural pressure. Confined aquifers may be replenished, or recharged by rain or streamwater infiltrating the rock at some considerable distance away from the confined aquifer. Groundwater in these aquifers can sometimes be thousands of years old.

#### Some related Aquifers

Two other common terms used are *Aquitard* (which retards groundwater flow) and *Aquiclude* (which excludes groundwater flow). These terms are also relative for the same reasons used as examples in the case of '*Aquifer*.' A number of similar or



synonymous terms exist for these features; aquicludes are also known as confining or impermeable layers, and aquitards as semi-confining or leaky impermeable layers.

### How does groundwater flow?

Gravity is the main force behind groundwater flow. However, there is a common misperception that groundwater flows in large subterranean channels, such as in the cave system. Groundwater flows mostly through the interconnected voids in rock. These may be the pore spaces between the grains in a rock, or cracks and fissures. The total volume of the pore space is known as the porosity. This represents the total volume of water that the rock can store. For the rock to be permeable, the void spaces must be interconnected, so that water can flow between them.

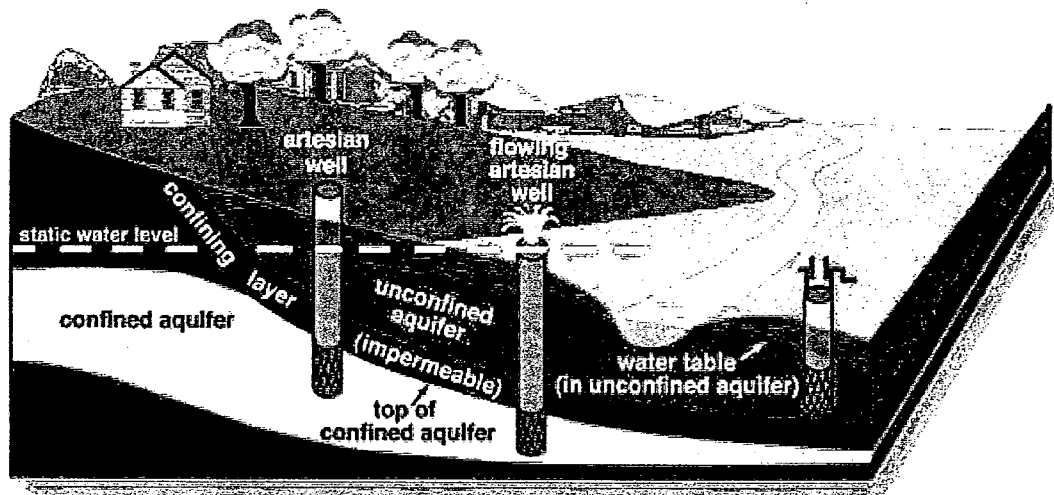


Fig.3- Different types of Aquifers (Kansas Geological Survey)

### Groundwater flow pattern:-

Groundwater can flow in different ways depending on the type and structure of the rock. The rate of groundwater flow, from springs or into boreholes, depends partly on the type of rock making up the aquifer. Flows can range from a tiny trickle out of sandy clay, for example, to thousands of cubic metres a day from some limestone aquifers.

Intergranular flow occurs when water moves between the grains in rock, for example in a sand or a sandstone. This is usually fairly slow. However, in limestones, cemented sandstones and many 'hard' rocks such as granites, most flow is along cracks and fissures. This is called fissure flow and is usually significantly faster than intergranular flow. In many aquifers there is no simple division between intergranular

and fissure flow. Both flow mechanisms can be present and play a greater or lesser part in overall groundwater flow. Water flows quickly in the fissures between the rock mass, but intergranular flow in the matrix is relatively slow.



Fig.4- Diff. Pattern of GW movement(GW Protection Framework)

### Groundwater flow concepts

The hydraulic gradient is the slope of the water table. It governs the direction of groundwater flow. The volume of flow through an aquifer is related to the hydraulic gradient and the hydraulic conductivity or permeability of the rock (a measure of how well pore spaces are interconnected). The speed of flow is related to hydraulic conductivity, hydraulic gradient and effective porosity (the connected void space in the rock). The speed of flow is given by the Flow Velocity.

In most aquifers, groundwater flow is slow. Speeds range from one metre per year to one metre per day. Occasionally, for example in highly fissured or karstic limestones, flow rates can be similar to those in rivers – in the order of kilometres a day.

### Darcy's Law

It relates the volume of discharge through an aquifer to the hydraulic gradient and the hydraulic conductivity

$$Q = k i a$$

$Q$  = discharge ( $\text{m}^3/\text{d}$ ),  $k$  = hydraulic conductivity ( $\text{m}/\text{d}$ ),  $i$  = hydraulic gradient,  
 $a$  = cross sectional area of flow ( $\text{m}^2$ )

*Flow velocity*

$$v = k i / n$$

$v$  = velocity in  $\text{m}/\text{d}$

$n$  = effective porosity

Groundwater discharges moves downstream (downhill). Such streams, referred to as winterbournes (or simply bourns), may remain dry for extended periods during droughts. These are natural seasonal variations. River flow can also be affected by groundwater abstraction. The relationship between the volume and timing of groundwater abstraction and river flows is complex. Inputs from urban runoff and sewage treatment works further complicate the situation and can hide natural inputs from groundwater. Sewage discharges can also hide inter-catchment transfers of groundwater with abstraction taking place outside the catchment where the discharge occurs.

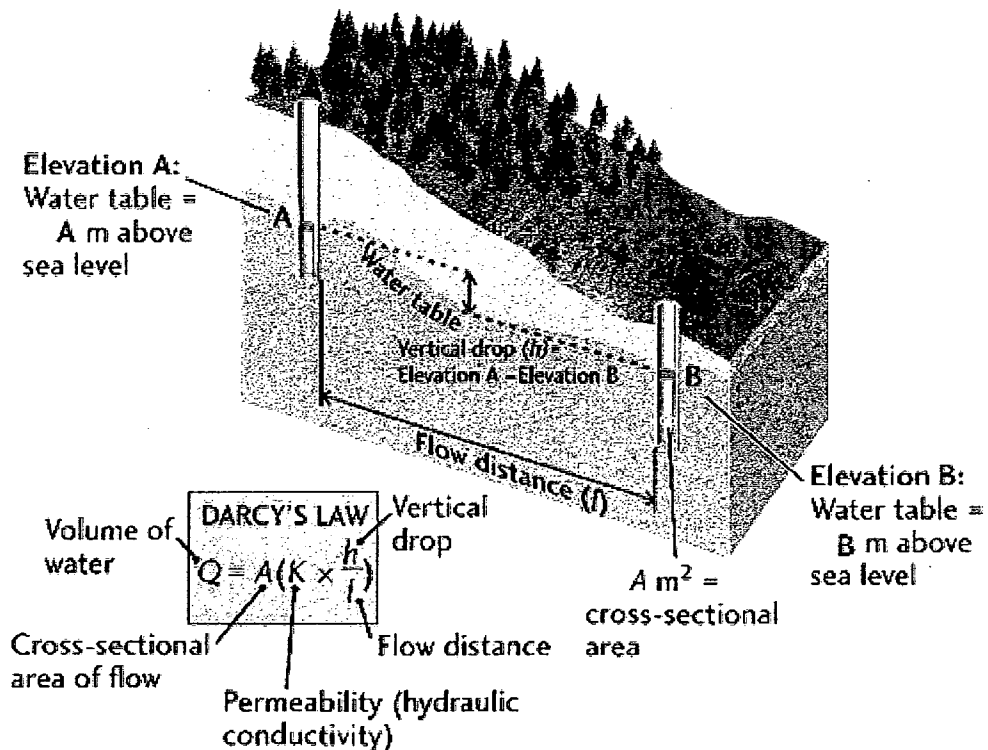


Fig.5- Pictorial diagram of Darcy Law ( GSCI 340)

**Groundwater quality standard (Given by WHO 1996)**

Parameter	Parametric value (mg/L)
pH	$\geq 6.5$ and $\leq 9.5$
Colour	Acceptable to consumers and no abnormal change
Taste	Acceptable to consumers and no abnormal change
Conductivity	2500 $\mu\text{S}/\text{cm}$ @ 20°C
Turbidity	Acceptable to consumers and no abnormal change
Sodium	200 mg/L
Calcium	200 mg/L
Magnesium	100 mg/L
Fluoride	1.5 mg/L
Chloride	250 mg/L
Nitrate	50 mg/L
Sulfate	250 mg/L
Ammonium	0.50 mg/L
Iron	0.2 mg/L
Manganese	0.05 mg/L
Arsenic	0.01 $\mu\text{g}/\text{L}$

Table (a) - Safe Drinking Water Standard prescribed by WHO (1996).**About Gangetic Plain of India:-**

The Gangetic Plain of India, with an area of about 700,000 square kilometers, varies in width by several hundred miles. It is the world's most extensive tract of uninterrupted alluvium. These deep, river-deposited sediments give rise to fertile soils. In addition, they are rich in groundwater for well irrigation. The second great structural component of India, the Indo-Gangetic Plain (also called the North Indian

Plain), lies between the Himalayas and the Deccan. The plain occupies the Himalayan foredeep, formerly a seabed but now filled with river-borne alluvium to depths of up to 6,000 feet (1,800 metres). The plain stretches from the Pakistani provinces of Sind and Punjab in the west, where it is watered by the Indus River and its tributaries, eastward to the Brahmaputra River valley in Assam state. The Ganges River basin (mainly in Uttar Pradesh and Bihar states) forms the central and principal part of this plain. The eastern portion is made up of the combined delta of the Ganges and Brahmaputra rivers, which, though mainly in Bangladesh, also occupies a part of the adjacent Indian state of West Bengal. This deltaic area is characterized by annual flooding attributed to intense monsoon rainfall, an exceedingly gentle gradient, and an enormous discharge that the alluvium-choked rivers cannot contain within their channels. Even so, to those who till its soils, there is an important distinction between *bhangar*—the slightly elevated, terraced land of older alluvium—and *khadar*, the more fertile fresh alluvium on the low-lying floodplain. In general, the ratio of *bhangar* areas to those of *khadar* increases upstream along all major rivers. The Ganga basin is joined by the river Yamuna from the west at Allahabad, then the rivers Ramganga, Gomti and Tons from the north and the Chambal, Betwa, Sinda and Ken from the south. In India the Ganges basin covers 8 states: Bihar (16.7%), Uttar Pradesh (34.2%), Himachal Pradesh (0.5%), Haryana (4%), Rajasthan (13%), Madhya Pradesh (23.1%), West Bengal (8.3%) and Delhi (0.2%). The catchment area of the Ganga basin is 1.06 million sq. km. out of which 861,404 sq. km. lies in India.

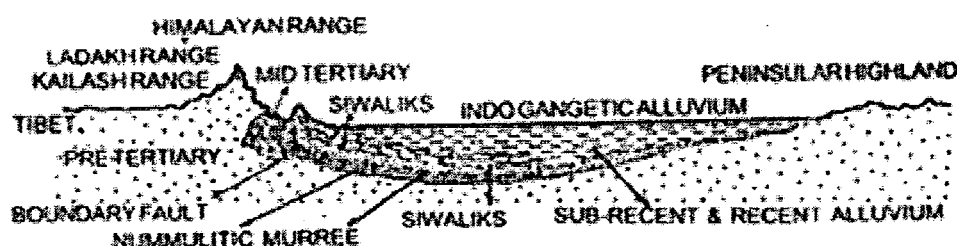


Fig.6- Cross section of Indo-Gangetic Plain (Tiwari 2003)

The rainfall distribution across the basin shows a general increase in average annual rainfall from east to west. The average annual rainfall ranges from 400-500mm from parts of Punjab to 1200 mm in West Bengal. Parts of the plateau highlands in the south also receive high rainfall as much as 2000 mm annually. Groundwater development is most suitable in the vast plains area of the basin.

Aided by development of canal networks that have induced recharge into the alluvial aquifers of the plains, rapid groundwater development has been experienced all across the plains areas in the past 50 years. The pattern of concentration of well density and availability of groundwater show variation across the plain areas. Freshwater parts of the western plains have shown high degree of development and there is extensive depletion of groundwater over vast areas, whereas saline groundwater areas in the western plains are showing signs of rising water tables. Towards the eastern plains, there is lesser intensive and profitability in exploiting groundwater, therefore, the degree of development is not as high. Population density is highest in the plain areas which are also marked by smaller land ownings.

In the plain areas, the rice-wheat cropping system is the most common. Productivity of agriculture is higher in the western areas as compared to the eastern areas per unit land and water applied. This can be attributed to various reasons. In general, the western regions are lesser dependant on rain-fed farming due to poorer rainfall. Therefore, there is greater reliance on irrigation from canal and from groundwater – both of which are relatively more reliable than the rains. This combined with the Warabandi system (fixed time period canal water allocation based on landholdings), better availability of energy for groundwater irrigation, natural factors such as lesser monsoon cloud cover (more sunny days per year) and being lesser prone to floods – are factors that have contributed to the relative economic prosperity of the western agricultural region. The presence of these local water markets combined with availability of tube well technology and energy availability (diesel, electricity) have contributed to the boom in groundwater development.

#### **Arsenic and its occurrence in groundwater:-**

Arsenic is present in small quantities in the Earth's crust. The element arsenic is 53<sup>rd</sup> in terms of abundance. Elemental arsenic has several allotropic forms - gray, yellow, and black arsenic- but only gray arsenic is ordinarily stable. Arsenic occurs naturally in soils, rocks, water, air, plants, and animals. Minerals that contain arsenic include arsenopyrite ( $\text{FeAsS}$ ), realgar ( $\text{AsS}$ ), orpiment ( $\text{As}_2\text{S}_3$ ), and arsenolite ( $\text{As}_2\text{O}_3$ ). The quantity of arsenic associated with lead and copper ores may range from 2-3%, whereas gold ores may contain up to 11% arsenic. Millions of years ago, as it drifted northwards, the Indian subcontinent collided with Asia. The resulting upward pressure created the Tibetan Plateau and the Himalayan mountain range. The region is drained by two rivers, the Ganges and the Brahmaputra, which subsequently join together to form a large, low-lying delta and alluvial plain. About

half of the territory of Bangladesh is situated in this river delta. One third of the delta lies in India, more specifically in West Bengal. It is estimated that something like 2.5 gigatonnes of sediment are carried downstream each year. The sediments contain a mixture of fine sand and clay. Their composition reflects that of the parent rock: large amounts of oxygen, silicon, iron and sulfur, but also trace elements such as gold and arsenic. The sediment carried down from the mountains, however, contains thousands of ppm of arsenic, which can bind to minerals in the soil or dissolve in water. This phenomenon is not restricted to Bangladesh. It also occurs in some areas of Argentina, Chile, India, Mexico, Taiwan, Thailand and the USA. Since 1998, a huge arsenic poisoning problem has been known to exist in Bangladesh. Naturally occurring arsenic commonly is found in volcanic glass in volcanic rocks of rhyolitic to intermediate composition; adsorbed to and coprecipitated with metal oxides, especially iron oxides; adsorbed to clay-mineral surfaces; and associated with sulfide minerals and organic carbon (Welch et al 1988). Sulfide minerals can contain arsenic either as a dominant mineral-forming element or as an impurity; sulfide minerals are found locally in the Western Cascades (U.S.G.S.1969).

#### **Arsenic cycle in the environment**

Major reactions in the soil-water and sediment-rock systems to influence the environmental transport, distribution and availability of arsenic. Oxygen availability controls the arsenate-arsenite redox reactions. Adsorption and precipitation of arsenate and arsenite immobilize the soluble arsenic. Slow release of arsenic from rocks and sediments or oxidative dissolution of arsenopyrite (FeAsS) from sediments contribute flux of arsenic in the environment. Methylation of arsenite to monomethylarsonic acid (MMA) or dimethylarsinic acid (DMA) followed by other organoarsenic compounds, constitute the major biological reactions in the arsenic cycle.

**Has arsenic over the years been released by the falling water table (due to the operation of numerous handpumps) and the penetration of oxygen or oxygen-rich water into the subsoil? Did certain soil bacteria play a role?**

Arsenic rarely occurs in free state, it is largely found in combination with sulphur, oxygen and iron (Brewster 1994; Chatterjee 1994). Arsenic occurs in the environment as a result of several inputs that contain this element as organic and inorganic forms (Rubio et al. 1992). The presence of arsenic in natural water is related

to the process of leaching from the arsenic containing source rocks and sediments (Robertson 1989; Hering and Elimelech 1995). Influx of arsenic from various anthropogenically induced sources may also contaminate both soils and ground water especially under anoxic conditions (Bhattacharya et al. 1996 & 1997).

The presence of arsenic in natural water is generally associated with the geochemical environments such as deposits of alluvial origin, volcanic deposits, inputs from geothermal sources, mining wastes and landfills (Welch et al., 1988; Korte and Fernando, 1991). Occurrence of arsenic in natural water depends on the local geology, hydrology and geochemical characteristics of the aquifer materials (Bhattacharya et al. 1997). The redox conditions in the subsurface environment also play an important role in determining the mobility of arsenic (Robertson 1986). The oxidation of different mineral species causes arsenic to become soluble and enter into the surrounding environment through drainage water. Any phenomena which affect the redox conditions such as pumping rate and the land use pattern is of interest in ascertaining the primary mechanisms responsible for the excess arsenic content in ground water. Robertson (1989) has further reported that occurrence and origin of arsenic in ground water depends on several factors such as adsorption-desorption, precipitation-dissolution, oxidation-reduction, ion-exchange, grain size, organic contents, biological activity and aquifer characteristics.

In the Bangladesh soil arsenic is chemically bound to sand, clay or other soil particles. Arsenic is also absorbed on fine iron and manganese oxyhydroxides, which decompose slowly and release arsenic. The type of chemical binding and its solubility are determined by various factors, such as the oxidation number, humidity, acidity, the amount of available oxygen, etc.

The geochemical cause of the arsenic problem is still not fully understood, but pyrite oxidation appears to play a major role (Bhattacharya et al. 1997). Pumping lowers the water table so that oxygen-rich water can enter the soil which was previously saturated. The oxygen causes decomposition of pyrite and the included trace elements are released. Arsenic in the atmosphere comes from various sources. Volcanoes release about 3,000 tonnes per year and micro-organisms release volatile methylarsines to the extent of 20,000 tonnes per year, but human activity is responsible for much more than these natural sources (Ferguson et al. 1972). About 80,000 tonnes of arsenic per year are released from the burning of fossil fuels. Each year, as much as 100,000 tons of arsenic is produced worldwide, with most of it



obtained as a by-product of the smelting of copper, lead, cobalt, and gold ores. Most rivers and streams contain arsenic concentrations less than  $1 \mu\text{gL}^{-1}$ . Oxygen-rich ground water can leach arsenic from arsenopyrite or other minerals and in this way the element can contaminate aquifers ( Ravenscroft et al. 2005).

### **Review of Geochemical Processes controlling Arsenic Mobility**

Two categories of processes largely control arsenic mobility in aquifers: (1) adsorption and desorption reactions (2) solid-phase precipitation and dissolution reactions (3) Oxidation of Arsenic Pyrites or Ferrous Hydroxides and (4) Oxy-hydroxide Reduction.

Arsenic adsorption and desorption reactions are influenced by changes in pH, occurrence of redox (reduction/oxidation) reactions, presence of competing anions, and solid-phase structural changes at the atomic level. Solid-phase precipitation and dissolution reactions are controlled by solution chemistry, including pH, redox state, and chemical composition. But the last two theories are most suitable in our area.

### **Mechanism of Oxidation of Arsenic Pyrites or Ferrous Hydroxides**

According to Singh AK (2006), arsenic rich arsenopyrite and ferrous hydroxide generally stable in a reducing environment (under the water table). It remains concentrated in organic deposits

But when oxidized, arsenic is released and adsorbed onto iron hydroxide. Main causes of oxidation are:-

A) Lowering of the water table below the organic deposits.

B) Result of dry seasons, pumping of wells, and dams built in the 1970's.

Poisonous Arsenic gets released during recharge, the arsenic adsorbed onto iron hydroxide returns to the reduced environment under the water table and is reduced to a toxic form.

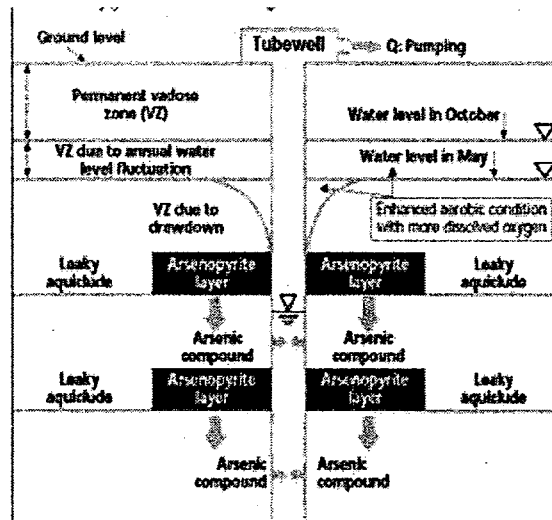
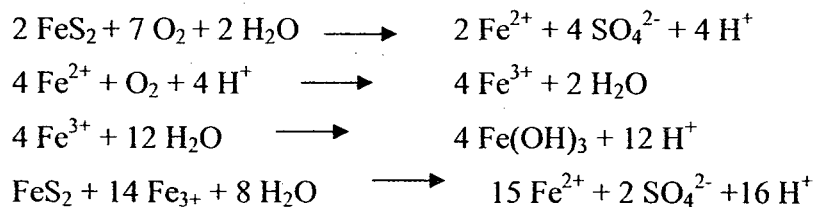


Fig. 6 (a) Release of As through Oxidation theory (www.inchem.org)

### Other possibility

By oxidation of Iron Pyrite which found in sediment layers formation of oxides of iron, sulfuric acid and arsenic takes place. But this form of arsenic is non-toxic. During recharge, sediments return to the reduced environment under the water table. Non-toxic oxides of arsenic are reduced to toxic forms (as shown in fig. 6(a)).



### Mechanism of Oxy- hydroxide Reduction

Organic matter deposited in the sediments reduces the arsenic adsorbed on the oxyhydroxides and releases arsenic into the groundwater when dissolution occurs during recharge. This is caused by microbial oxidation of the organic matter as bacteria dissolve surrounding oxygen. Arsenic is naturally transported and adsorbed onto fine-grained iron or manganese oxyhydroxides (Singh AK, 2006). Highest arsenic concentrations are usually found in fine-grained sediments. Aerobic bacteria activities oxidize arsenite to arsenate.



### Arsenic speciation and its importance in study

The speciation of an element is the distribution of an element amongst defined chemical species in a system. The chemical species are specific forms of an element defined as to isotopic composition, electronic or oxidation state, and/or complex or molecular structure. Florence (1982) has defined the term speciation analysis as the determination of the individual physico-chemical forms of the element, which together make up its total concentration in a sample. The knowledge of the geographic distribution of different arsenic species in natural water systems is important for environmental consideration of the geochemical and biological cycling of the element Jakariya et al. (2007). Furthermore, this will also provide insight into the geochemical process responsible for elevated arsenic concentrations in different hydrogeological environments.

It is essential to understand the levels of As in drinking water and its speciation to develop regulatory standards. In natural waters, As is typically occurs as weak triprotic oxyacids. In reducing environment, arsenous acid dominates in the form of  $\text{H}_3\text{As}^{3+}\text{O}_3^0$  at a wide range of pH values while the protonated  $\text{H}_3\text{As}^{3+}\text{O}_3^-$  forms only at  $\text{pH} > 9.0$ . At higher pH and in an oxidized environment, As v is present as  $\text{H}_2\text{AsO}_4$  ( $\text{pH} < 7.0$ ) (Cotton and Wilkinson 1988). According to the following equation shown in fig. 6(b) arsenic acid is a moderately strong oxidizing agent and is readily reduced to arsenous acid (Bhattacharya et al 1999).

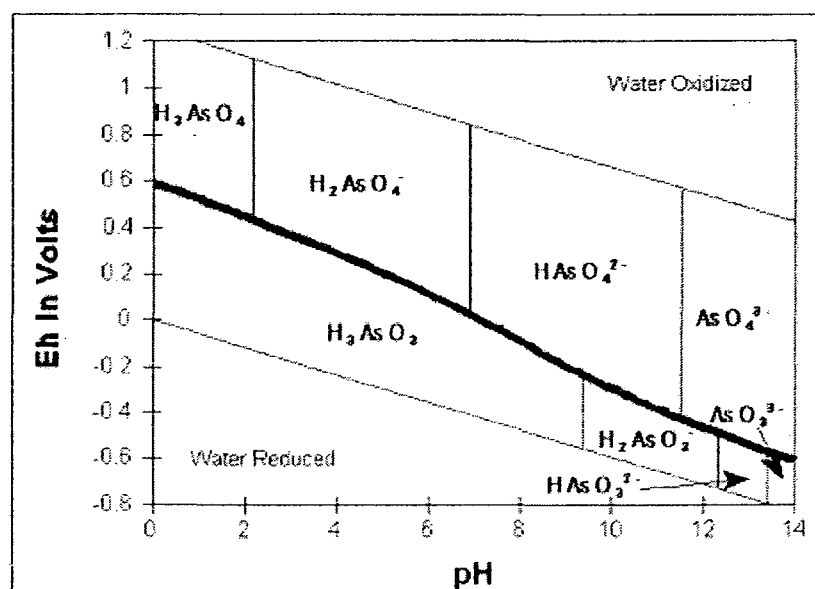


Fig. - 6 (b) Eh/Ph conditions and arsenic speciation

([www.inchem.org/documents/ehc](http://www.inchem.org/documents/ehc))

Arsenic exists in the environment in a number of valency states. The valency state of arsenic plays an important role for its behavior and toxicity in the aqueous system. The toxicity and bioavailability of arsenic can only be determined if all its forms can be identified and quantified. Current interest in the determination of different species of arsenic in natural waters also because of knowing the fact that physiological and toxic effects of arsenic are connected with its chemical forms. The toxicity of different arsenic species varies in the order: arsenite > arsenate > monomethylarsonate (MMA) > dimethylarsinate (DMA) (Penrose 1974; Lewis and Tatken 1978; Stugeron et al. 1989). The concentration of arsenic in natural waters depends on the geological composition and the degree of pollution of the environment. The concentration of As(III) to As(V) varies widely depending on the redox conditions in the geological environment (Braman and Foreback 1973; Andreae 1977; Shaikh and Tallman 1978). The toxicities of various arsenic compounds to man and animals have been studied by various workers (Sullivan 1969; USEPA (United States Environmental Protection Agency) 1971; Fairchild et al. 1977). Chatterjee (1994) has discussed the toxicity of arsenic to different organs. Biologically, As(III) is considered more toxic than As(V) (NAS (National Academy of Sciences) 1977). Data on the differences in toxicity between As(III) and As(V) on human beings are very limited. Trivalent arsenic is about 60 times more toxic than oxidized pentavalent state (Ferguson and Gavis 1972). Inorganic arsenic compounds are about 100 times more toxic than organic arsenic compounds (DMMA and MMAA) (Nagy and Korom, 1983).

### **Biological Toxicity**

The toxicology of arsenic is a complex phenomenon as arsenic is considered to be an essential element also. Two types of toxicity, viz., acute and sub-acute are known from long time. The acute arsenic poisoning requiring prompt medical attention usually occurs through ingestion of contaminated food or drink. The major early manifestation due to acute arsenic poisoning includes burning and dryness of the mouth and throat, dysphasia, colicky abnormal pain, projectile vomiting, profuse diarrhea, and hematuria. The muscular cramps, facial edema and cardiac abnormalities, shock can develop rapidly as a result of dehydration (Done and Peart 1971). Sub-acute arsenic toxicity mainly involves the respiratory, gastro-intestinal, cardio-vascular, nervous and haematopoietic systems. It may cause loss of appetite, nausea and some vomiting, dry throat, shooting pains, diarrhea, nervous weakness,

tingling of the hands and feet, jaundice and erythema. Longer exposure resulted in dry, falling hair, brittle loose nails, eczema; darken skin exfoliation and ahorny condition of the palms and soles (Holmquist 1951; Pinto and McGill 1953).

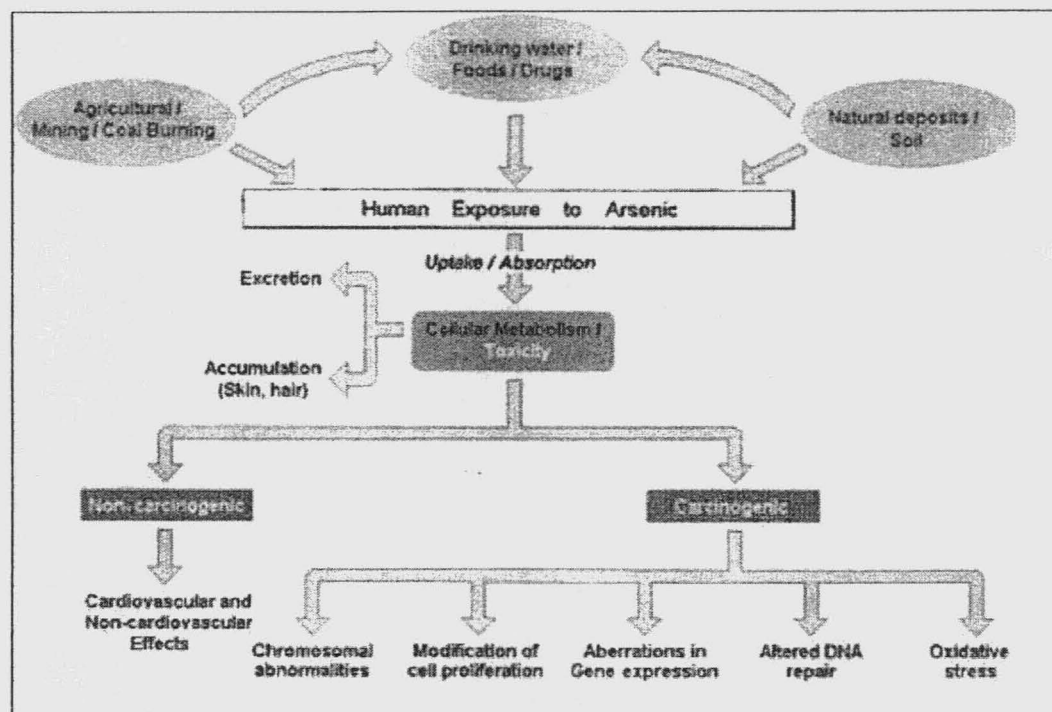


Fig. -7. Sources of Human Exposure to Arsenic and various modes of arsenic toxicity (www.devalt.org/newsletter)

### Why I have taken this area?

Ground water with high concentration of dissolution (more than recommended by WHO (1996) as safety limit) in the sedimentary aquifers in Southeast Asian countries is the chief source of chronic human intoxication (Bhattacharya et al. 1996). So the major concerns are to identify chemical and physical characterized ions, and its compounds from natural and anthropogenic processes which interact with living system. Besides natural sources, increasing anthropogenic activities like land use changes, overexploitation etc. are also responsible for enriched dissolution in ground water.

Arsenic mobilization in ground water is considered to be reflection of redox geological environment present their along with the cumulative effect of anthropogenic processes happen in the main river and its tributaries in and around Gangetic plain. It is so prime importance to identify different mechanism of arsenic mobilization, transportation and its enrichment in ground water to know its present condition and to find out ways to check its mobility and toxicity.

**Objective**

Published literature reports indicate availability of detailed scientific study on arsenic in groundwater of Gangetic plain are very limited, so this study has been attempted with the following objectives:

1. Determination of groundwater quality of two districts in space and time.
2. Geochemical characterization and water quality classification etc to find out suitability of water for drinking /domestic/ irrigation purpose by targeting safe aquifers.
3. Statistical analysis of the water quality data.
4. Distribution of arsenic in shallow and deep aquifer in both districts.
5. To quantify arsenic in groundwater and identification of possible polluted regions.
6. Arsenic speciation ( $As^{+3}$  and  $As^{+5}$ ) in ground water of study area to identify the toxicity level.

**Relevance**

The study will identify the distribution, mobilization of arsenic in groundwater of both districts (Bhagalpur and Ghazipur) in central Gangetic plain. The outcome of study will help to delineate the arsenic polluted zone and help to target safe aquifers for drinking / domestic / irrigation purpose. The outcome will help to adopt suitable mitigation for removal; remediation to suggest safe drinking water purpose in these two districts. This will be helpful to generate awareness to avoid mass casualty in near future.

The geochemical properties of groundwater also depend on the chemistry of water in the recharge area as well as the different geochemical processes that are occurring in the subsurface. These geochemical processes are responsible for the seasonal and spatial variations in groundwater chemistry (Matthess 1982). Groundwater chemically evolves by interacting with aquifer minerals or internal mixing among different groundwater along-flow paths in the subsurface (Domenico 1972; Wallick and Toth 1976; Toth 1984).

The hydro-geochemical character of groundwater and its quality in different aquifer over time and space proved to be an important technique in solving the problem (Chebotareb 1955; Hem 1959 & 1991; Gibbs 1970; Chandrashekharan et al. 1988; Jankowski and Jacobson 1989; Jain 1996 & 1997; Atwaria et. al. 1997; Ballukraya and Ravi 1999; Anandhan et. al 2000). Hydro geochemical evaluation and relative studies were done in detailed by Back and Hanshaw (1965).

Water chemistry can vary tremendously over very short distances depending upon whether a sample is drawn from an actively flowing conduit or from rock matrix (Quinlan and Ewers 1985). Water chemistry also varies over time in response to seasonal changes in recharge and dilution effects of individual stream (Hess and White 1988). Along subsurface flow path, water can be modified by CO<sub>2</sub>, mineral dissolution and/or precipitation, and mixing (Dreybrodt 1981; Herman and Lorah 1986; Holland et. al. 1964).

The hydro geochemical interpretation of the data obtained and their graphical representation for meaningful interpretation was put forth by various diagrams like Piper diagram, Durov plot, Schoeller diagram (Piper 1944; Durov 1948; Schoeller 1965). The chemical concentration of various ions in absolute and relative values will be used in the interpretation of chemical evolution of groundwater and to observe salinity. Highly saline and brackish groundwater in the north-western and south-western parts of the area seems to be associated with long history of evaporation and to oxidation of sulphur gases in low lying areas (Datta and Tyagi 1996).

Abu-Taleb (1999) studied in detail the infiltration studies and artificial recharge. Aquifer recharge studies using different isotopes helping determination of infiltration capacities, permeability and other aquifer parameters. This technique was adopted for hydro-geological studies (Singh et. al 1999). Significant quantities of evaporated surface run-off water and rain water infiltrate along with nitrate and fluoride salts in the soil (Datta et. al., 1999).

In the study area Delhi, experiments carried out by Kakar (1985) and Kumar et al, (2006) showed that salinity is in patches, which cannot be caused by airborne salts. Using selected chemical data, Datta et. al (1996) found that the local recharge is associated with low salinity water of Ca-Mg-HCO<sub>3</sub> type. Highly saline and brackish ground water in the northwestern and southwestern parts of the areas has long history of evaporation and oxidation of sulphur gases in low lying areas (Datta et. al 1996).

The salinity problem in groundwater is increasing in coastal areas including some part of Haryana, Rajasthan, Uttar Pradesh and Punjab state. The leakage in Jhamarkota phosphorites mines in Rajasthan has been also studied by Kulkarni et al. (1998) using hydrochemistry of groundwater and environmental isotopes. It was found that recycled water reaches mines through the highly permeable geologic formation in the vicinity of the mines.

Identification of recharge sources and location of recharge zone are one of the most important areas of studies that needs immediate attention of the scientific community to save the zone of recharge of deeper aquifers from the anthropological activities and contamination. It has been observed that in most of the cases, the details about the recharge resources and location of recharge zones for deeper aquifers are not available in India. The information about interconnection is also very useful to understand the availability of groundwater in different aquifers and to take necessary measures for saving aquifers from contamination. But, it seems that no sincere efforts have been made in this direction to study aspects of different aquifer system in the country (Rao et. al. 2000).

Shallow rural domestic wells are most likely to be contaminated with nitrate especially in the areas where nitrogen based fertilizers are used. Other sources include disposal of sewage and industrial waste, and handling of animal's manures. A number of researchers have reported on the nitrate contamination in the various aquifers (Singh and Sekhon 1978; Jacks and Sharma 1983; Tesoriero and Voss 1997; Keshari 1999 & 2003) carried out a study for prognosis of nitrate distribution in the Faridabad- Ballabhgarh aquifer. They found that nitrate concentration decreases with depth of water table, and animal wasters are the major contributor to the nitrate level in groundwater near village area. Local disturbances that affect nitrogen cycling can lead to large nitrogen losses as nitrate to groundwater (Keeney 1980; Frazer et. al. 1990; Dillon et. al. 1991).

For Piper Diagram, groundwater is treated as though it contained three cations constituents (Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup> + K<sup>+</sup>) and three anions constituents (F<sup>-</sup> + Cl<sup>-</sup> + NO<sub>3</sub><sup>-</sup>,



$\text{SO}_4^{2-}$ ,  $\text{HCO}_3^-$ ) expressed as percentages in milli-equivalent per litre of cation and anion respectively ( Todd 1980).

Most of the groundwater samples tend to show a charge imbalance mainly in favour of positive charge excess, but some inversely with a negative charge deficit. Positive charge excess higher than 5% agrees with the database of the dissolve load. This imbalance of negative charges could be related to the fact that no analysis was of organic matter (Berner-kay and Berner 1987; Edmond et. al. 1995; Huh et. al. 1998), which is mainly produced by biological activities during summer and spring.

In the hydrological evaluation, if the evaporation is dominant, assuming that no mineral species are precipitated, the Na/Cl ratio would be unchanged. If halide dissolution is responsible for sodium, the Na/ Cl molar ratio should be approximately equal to one, whereas a ratio greater than one is typically interpreted as Na released from a silicate weathering reaction (Meyback 1987).

If the ratio Ca/ Mg is equal to one, dissolution of dolomite should occur, whereas a higher ratio is indicated of greater Calcite contribution. Higher Ca/Mg molar ratio  $>2$  indicate the dissolution of silicate minerals, which contribute Ca and Mg to groundwater (Llyod and Heathcode 1985).

Manchanda (1976) provided block wise groundwater quality details for the state of Haryana. The contribution of poor groundwater quality towards total groundwater development in arid and semi arid region of India is a proximately 32 to 84% (Minhas and Gupta 1992) in quality regions, the average extent of saline, sodic and saline-sodic water are approximately, 20, 37 and 43% respectively (Yadav and Kumar 1995). A chloride concentration of 600 mg/l has been considered as the highest acceptable salinity level for human consumption (Rosenthal 1994).

Bicarbonate is mainly derived from rock weathering and the pollution contribution is minimal ( Meybeck 1979). The groundwater can be classified based on the TDS concentration as fresh water 0-1000 mg/l; brackish water 1000-10,000 mg/L; saline water 10,000-100,000 mg/l (Freeze and Cherry 1979). Groundwater flowing through a normal and active hydrological gradient is characterized by Na/Cl ratio of 0.86-1.00 (Rosenthal 1994). The higher ratio than normal ratio indicates no/lesser mixing of seawater and fresh water within the aquifer.

In recent times the interest in fluoride has greatly increased, owing to its importance in the precipitation of fixation of phosphate in mineral like fluoro-apatite and to the recognition of pathological condition in man and animals, described as fluorosis (Agarwal et. al. 1997; Rao et. al. 1998). Under acidic circulation, solution

fluoride is readily absorbed in clay structure, while in alkaline environment; it is desorbed (Deshmukh et. al. 1995).

Nigam (1999) investigates the hydro geochemistry of fluoride in ground water of Agra district (U.P.). The study shows concentration of fluoride ranges from 0.09 to 7 mg/L and fluoride is showing good correlation with Bicarbonate ion. The high concentration of fluoride in groundwater of the study area is due to rock water interaction i.e. between groundwater and easily weathered fluoride bearing mineral like fluorite, fluor-apatite etc. High fluoride in groundwater samples in Gangetic plain have been reported by Chadha and Tamta (2001) for some villages in Unnao district in Uttar Pradesh.

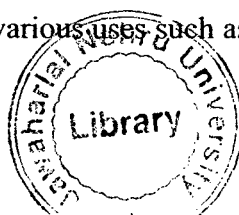
Sub-surface water constitute of water present in the unsaturated zone (vadose zone) known as soil moisture and water present below the water table i.e. in saturated zone known as groundwater. Results reported by Laney (1978) and by McQueen and Miller (1972) showed that water in the unsaturated zone may have the relatively high solute concentrations with the underlying water.

Initial studies in groundwater mainly related with the effect of lithology on the water composition. Kimmel and Braids (1980) worked on the hydrochemistry of quartz sand aquifer on long island, Freeze and Cherry (1979) investigate the change in the composition of groundwater as it moves through aquifer due to addition of constituents.

Shanyengana et. al. (2003) investigated the major ion chemistry and ground water salinization in ephemeral flood plains in some arid regions, according to them the dominant process occurring in the area which determine the hydrochemical shift are refreshing by recharge water , concentration by evaporation, dissolution of saline sediments (mainly evaporates) mixing with older and more saline groundwater.

Rugge et. al. (1992) indicate that concentration of organic compound can be very high in the landfill leachates that can have effect on the oxidation-reduction potential of groundwater, and indicate that it may decrease the oxidation-reduction potential(ORP), which can mobilize toxic metals. Kelly et. al.(1996) indicate that soil in industrial areas are commonly contaminated with heavy metals, which may potentially leach into shallow groundwater. The calculation of mineral saturation index and thermodynamic equilibrium studies were initiated by Garrel and Christ (1964). Similar works were done by many workers (Back 1963; Krauskopf 1983).

In India ground water research is mainly focus toward analyzing groundwater for determining its suitability for various uses such as irrigation purpose, water supply



for public use etc. Jacks (1983) investigated the effect of crop farming on groundwater quality of Coimbatore district of Southern India. The area is comprises of igneous rocks. Groundwater is used for irrigation purpose so it often subject to intense evapotranspiration amounting to 600-1300 mm per harvest. High salinities are caused by the evapotranspiration together with the hydrological factor.

Tamta (1999) studied the occurrence and origin of ground water salinity in Bhatinda District, Punjab. The study indicates presence of salinity is largely due to simple dissolution of minerals in the aquifers, low ground water flow, mixing of ground water and infiltration of evaporated irrigation water along with fertilizers  $\text{NO}_3^-$  and  $\text{K}^+$  to ground water system.

Som and Bhattacharya (1992) investigate the groundwater geochemistry of recent weathering at Panchpatimali Bauxite bearing plateau, Koraput district, Orissa. During their study they found that ground water is feebly acidic with positive oxidation potential which transform  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$ , Kaolinite is the main weathering product with marginal development of gibbsite. Relative mobility of Al is nil, Fe low and Mg, Ca and Na concentration is high which are important for the development of aluminium silicate and iron oxide/hydroxide.

Pawar (1993) investigate geochemistry of carbonate precipitation from the groundwater in the basaltic aquifer, he found that most groundwater are saturated with respect of calcite and dolomite. The molar Ca/Mg values are less than unity in most of the samples and value more than unity for Na/Ca supports evidence for the precipitation of calcite particularly in monsoon period.

Rao et. al. (1998) studied the impact of industrial effluent on ground water quality of Venkatapuram area, Visakhapatnam, Andhra Pradesh. Study indicates that leakage of surface water from the effluent tank and along the stream course forms a major source of groundwater contamination.

Pawar and Nikumbh (1999) studied the trace element in the Behedi basin, Nasik district. Their study reveals that the trace metal like Fe, Mn, Cu, Zn have entered into the aquifer due to rain fed recharge. They attributed the spatial variation due to lithologic and anthropogenic sources, such as application of fertilizers and pesticides. Mehrotra and Mehrotra (1999) studied the pollution of groundwater by Mn in Hindon-Yamuna Doab district, Ghaziabad. Anthropogenic activities coupled with natural hydro geochemical environment in the area are responsible for high metal concentration in the area.

## **Arsenic related literature review**

### **World wide arsenic distribution scenario in ground water**

Arsenic is a toxic element has been reported in many countries, including Vietnam, Mangolia, Thailand, Argentina, Bolivia, Chili, Taiwan, India, Bangladesh and many others. Natural geo-chemical mechanisms are controlling the groundwater chemistry in the aquifers of the Mekong delta region of Vietnam (Smith et al, 2000). As in the ground waters is a range between 1-3050  $\mu\text{g/L}$  (average 159  $\mu\text{g/L}$ ). It suggests that As concentrations are not high in the ground waters, though in some places it has been found with concentrations > 50  $\mu\text{g/L}$  further upstream in the Mekong Valley of Cambodia. Choprapawon and Rodcline (1997) reported that in the Ron Philbun district of Malaysia the wells are polluted with As, its concentration is up to 6700  $\mu\text{g/L}$ . Recent findings of arsenic in the Tarai area of Nepal has been highlighted by Chakraborti et. al. (2003). Incidences of chronic arsenicosis due to consumption of arsenic contaminated water in different parts of India were gradually increasing since 1983. This is the first specific information regarding arsenic distribution of groundwater in SW Uruguay. Twenty-eight wells were sampled on the aquifers of Mercedes, Raigon and Chuy in five localities (Manganelli et al 2007).

### **Arsenic distribution in the Indian sub-continent groundwater**

Natural As contamination in ground waters of the Bengal Delta Plain (BDP) is prominent, covering the state of West Bengal, adjoining country of Bangladesh and now it is extended to Bihar, Jharkhand, Uttar Pradesh, Punjab, Chattisgarh, east coast of India (Chennai), western and eastern India and north-eastern India, and the neighboring country of Nepal towards north of the Indo-Gangetic Alluvium (Cotton et al. 1998; Cullen et al. 1989; Datta et al. 2000; Ferguson et al. 1972; Fordyce et al 1995; Huq et al 2000; Lund U., Fobian A. 1991; Madhavan N, Subramanian V, 2000). Geologically, the Bengal basin is situated in the region of intense neotectonic activity (Mandal, NK, Biswas R 2004) and its west, north and east are bordered by the Indian Shield, Shillong Plateau, and Naga-Lusai orogenic belt, respectively. The BDP which is prograding delta opens out to the Bay of Bengal towards south (Mukherjee et al., 2001). The considerable amount of As is released to groundwater by the sediments deposited by the rivers during the late Quaternary or the Holocene age. Lithology of these late Quaternary sediments includes sands, silt and clay; mineralogical composition of these sediments consists of quartz, feldspars, illite and kaolinite, and

the fine-grained overbank facies are rich in organic matter (Naidu R. 2000; Nickson et al 2000; Nickson et al 1998; Pandey et al., 2002).

#### **Arsenic scenario in South and west India**

Ramesh and Ramanathan (2002) studied the groundwater quality of Chennai city and reported that at certain pockets of the city in groundwater As concentration is high. This is because of industrial pollution. Significant concentration of As was observed near Adyar mangrove creek (<10 µg/L to 146 µg/L), this is because of concentration of As is due to anthropogenic pollution, the paint, textile, tanning and chemical manufacturing industries, as well as due to the geogenic sources.

High concentration of As was observed in the ground water is reported by Madhavan and Subramanian (2000) in area around the Khetri mines in Jhunjhunu district and Zawar mines in Udaipur district of Rajasthan.

#### **Arsenic scenario in East and North-east India**

Chidambaram et al. (2006) studied the mineralogical control over the As distribution in the east coast groundwater. Nearly 50 million people living in 3200 villages of 9 of the total 18 districts of West Bengal exposed to drinking water containing As 10 µg/L (Ramesh et al. 2002; Roychowdhury et al 2003; Singh et al 1991). The pH and Eh of the Bengal ground water is ranged between 6.5 to 7.6 and +0.59 to -0.44 V. The generally the water is of Ca-HO<sub>3</sub>, Ca-Mg-HCO<sub>3</sub>, although Ca-Na-HCO<sub>3</sub> type and Na-Cl type water are also observed in certain pockets of the Bengal plain (Nickson et al., 2000; Singh, 2004). Mukherjee and Bhattacharya (2001) and Roychowdhury et al (2003) indicated that high DOC levels are consistent with As (III) in ground water which suggests that reduction of organic matter by microbes and conversion of As (V) to As (III) in the sedimentary aquifers responsible for the higher levels of As contamination in the Bengal ground water. The predominance of arsenic poisoning due to contaminated groundwater in West Bengal has been thought to be limited to the Ganges delta (the lower Ganga plain). Early surveys have been conducted on arsenic contamination in groundwater of West Bengal (Saha 1984; Mazumdar et. al. 1998). The arsenic contamination in the groundwater particularly in the alluvium plain of Gangetic plain may cause secondary effect as well which result from agricultural activities and ultimately enters the human food chain. High arsenic in the groundwater of lower Gangetic plain of West Bengal was reported by (Bhattacharya et. al. 1997).

In the northeast region of India, the regions of Assam and Tripura which are close to Bangladesh, where the concentration of As has been found to be above 300

$\mu\text{g/L}$  where as in Manipur As is in the range between 798-986  $\mu\text{g/L}$ , in Arunachal Pradesh 58-618  $\mu\text{g/L}$ , in Tripura 65-444  $\mu\text{g/L}$ , in Nagaland 50-278  $\mu\text{g/L}$  (Madhavan et al 2000). It is believed that the weathering of sulfide associated with carbonaceous matter may have produced As rich iron oxyhydroxides which in turn released As (after reduction) to the existing sedimentary environment (Chowdhury et al 2000).

The study area is located on the western part of the alluvium-filled gap between the Rajmahal hills on the west and the Garo hills on the east. Groundwater occurs under unconfined condition in a thick zone of saturation within the Quaternary alluvial sediments. Three hydrochemical facies with distinct characteristics have been identified which are dominated in general by alkaline earths and weak acids. The major-ion chemistry of the area is controlled by weathering of silicate minerals (Sikdar and Chakraborty 2007).

### **Arsenic scenario in North India**

In northern Indian states (Bihar, Jharkhand and Uttar Pradesh), considerable load of As is present in ground water. Chakraborti et al. (2003) in Bhojpur and Buxar districts of Bihar As concentration groundwater in 107 out of 152 villages exceeded the WHO and the national drinking water standards. 6809 samples of groundwater collected from hand pumped tubewells, in which 42% of the wells containing As concentrations  $> 10 \mu\text{g/L}$ , 24% of wells had concentrations of As  $> 50 \mu\text{g/L}$  and about 6% wells contained As  $> 300 \mu\text{g/L}$  Groundwater As contamination has also been detected in the state of Jharkhand, situated on the western bank of river Ganges. Groundwater in 'the district of Sahibgunj district containing As concentration more than WHO and national drinking water standards. Chakraborti et al. (2003) analyzed groundwater samples from 25 villages of Uttar Pradesh and found that in 914 hand tubewells As concentration was  $> 50 \mu\text{g/L}$  in 22 villages. Acharyya et al. (2005) reported that in Chattisgarh state the highest concentration of As was observed in Sansayatola village of Ambagarh-Chowrin block (avg. 436 $\mu\text{g/L}$ ) than Kaurikasa (avg. 244 $\mu\text{g/L}$ ), Jadutola (avg. 104  $\mu\text{g/L}$ ) and Dhadutola (10  $\mu\text{g/L}$ ).

Arsenical skin lesions and hyperkeratosis marks of a patient from Semria Ojha Patti village, Bihar, India was already reported in this state (Chakraborti et al. 2003). The occurrence of Arsenic in Upper and Middle Gangetic plain groundwater are fairly new because most of the documented cases are reported from Bengal basin of Bangladesh (Ahmed et. al. 2004; Ben et. al. 2003).

The As and related contamination in groundwater and surface water in Ballia district of Gangetic plain in northern India were reported by Tripathi et al. (2006) .

Arsenic concentration in Ballia varied from below detection limit up to as high as 200  $\mu\text{g/L}$ . The observed concentration of As was very high in the location close to the river basin. The As in groundwater in this district shows that intermediate aquifers have more As compared to Deep and Shallow aquifers. The effected aquifers seem to be particularly in risk, due to the prevailing geochemical conditions in which oxidized and reduced waters mix, where the amount of sulphate available for microbial reduction seems to be limited. The high arsenic concentration (100-180  $\mu\text{g/L}$ ) was in Belahari, Ramgarh, Reoti, & Bairyia regions of the middle Ganga-Ghaghra plain (Tripathi and Ramanathan et al 2006).

### **Recent literature review**

In the natural conditions, speciation of As changes qualitatively according to the thermodynamic principles (Chakraborti et al. 2004). In the As-H<sub>2</sub>O-O<sub>2</sub> system, stable inorganic As species are H<sub>3</sub>As<sup>III</sup>O<sub>3</sub>, H<sub>2</sub>As<sup>V</sup>O<sub>4</sub><sup>-</sup>, H<sub>2</sub>As<sup>V</sup>O<sub>4</sub> or As. However, in the presence of dissolved S, As sulfides (AsS<sub>2</sub><sup>-</sup>, As<sub>2</sub>S<sub>3</sub> and HAS<sub>2</sub>) are stable (Chidambaram S 2006).

Several authors suggested that reductive dissolution of Fe (III)- oxyhydroxides in strongly reducing condition of the young alluvial sediments is the cause of mobilization of arsenic (Ahmed et. al. 2004; Mc Arthur et. al. 2004).

Continued investigation by SoES-JU in eastern Uttar Pradesh has found that out of their total samples 46.5% of contain arsenic in concentrations >10 $\mu\text{gL}^{-1}$  and 26.7% of their samples have > 50  $\mu\text{gL}^{-1}$ . Arsenical skin lesions have been reported in people from Ghazipur and eastern U.P. district (Ahamed et al, 2006).

Elevated concentrations of As have been reported from Ganges–Gaghra Plain, Ballia district, eastern Uttar Pradesh (Tripathi and Ramanathan, 2006).

The organic carbon undergoes oxidative carbon degeneration by different oxidants and increases the concentration of CO<sub>2</sub> in the aquifer. The reducing condition thus developed in the aquifer helps to dissolve the arsenic adsorbed on iron hydroxide or oxy-hydroxide coated margins of sand, iron rich heavy mineral grain margins, clay minerals and Fe-Mn concretions present in the aquifer matrix (Sikdar and Chakraborty 2007).

Upstream of Bihar in Uttar Pradesh, to date water samples from 20,126 government-installed hand pump sources have been tested. As a result 2.4% of the samples tested were found to contain arsenic at concentrations greater than 50  $\mu\text{g L}^{-1}$  and 21.5% at concentrations greater than 10  $\mu\text{g L}^{-1}$  (Nicksion et. al, 2007).

The groundwater arsenic contamination and its health effects in different parts of India and south- East Asia has been critically reviewed by Ramanathan et al (2007). The distribution of As in shallow groundwater of the Bengal Basin is likely to be controlled by multiple processes. The new data from widely separated portions of the Bengal Basin are consistent with Fe oxyhydroxide reduction as a source of As, but also suggest other potential mechanism that may be important in relatively young deposits (Metral et al. 2008)

The postoxic main aquifer water exhibits overlapping redox zones (metal-reducing, sulfidic and methanogenic), indicative of partial redox equilibrium, with the possibility of oxidation in micro-scale environments. The redox processes are depth-dependent and hydrostratigraphically variable. Elevated dissolved As in the groundwater is possibly related to Fe (III) reduction, but is strongly influenced by coupled Fe-S-C redox cycles. Arsenic does not show good correlations with most solutes, suggesting involvement of multiple processes in As mobilization (Mukherjee et al 2008)

Above extensive literature reviews indicate that there is no study on general hydrochemistry and enrichment of arsenic in the groundwater of Bhagalpur and Ghazipur is available. Hence an attempt has been made in this study in two districts of Central and middle Gangetic plain.





In the field with Dr. AL. Ramanathan during sample collection and primary tests.



Water collection from Tube well as well as Arsenic Speciation in field

## **B**HAGALPUR

### **Location and Area**

Bhagalpur district is located in the eastern alluvial part (classification based on agro-climatic zonation) of the Bihar ranges from 25°07.97' to 25°36.48' N latitude and from 86°38.44' to 87°32.52' E longitude (Map1). According to the central statistical organisation the district had an area of 2,570 sq. km. on 2001. Bhagalpur is a city and municipal corporation in Bihar state in eastern India. The city had a big harbour on the Ganges River at place called Champanagar (another name for Bhagalpur) now called Champanala which flows next to western boundary of present city near Nathnagar. The gangetic plains are very fertile and the main crops include rice, wheat, maize, barley, and oilseeds. The economy of Bhagalpur is dependent mainly on agriculture and silk.

### **Demography**

As of 2001 India census, Bhagalpur had a total population of 19, 09,967 out of which rural portion occupies 15,66,518 and urban occupies 3,43,449. Males constitute 54% of the population and females are about 46%. Bhagalpur has an average literacy rate of 68%, with 70% of the males and 62% of the females literate.

### **Geomorphology**

#### **River system**

The district is a peneplain, intersected by numerous streams. Surface levels varied due to high banks of the Ganga, Koshi (Ghugri), Chanari, and Chandan. The geomorphology of the area is monotonously flat and featureless plain with gradient towards the river Ganges.

#### **The Ganges**

The Ganges enters the district opposite the village of Sultanganj, where a great mass of granite rises out of its bed. After crossing ranges of hills at the place called Patharghat, it receives the united water of Koshi and all the northern rivers of the district. Average width of the river is three miles but during the rainy season extra two miles on either side also get inundated.

#### **The Chandan**

The chandan is the largest hill stream in the south of district, which joins the river Ganga near Ghogha.

### **Climate and Rainfall**

The southwestern monsoon brings the much-needed rainfall and nearly 70 % of the rainfall is received during South-West monsoon months i.e. July to September. There is significant seasonal variation in temperature with mean monthly temperature being as high as 45 °C in June and as low as 2 °C in January while mean monsoon temperature is 24 °C. The major part of the district qualifies for arid soil moisture regime according to the criteria laid in Soil Taxonomy. The climatic condition of this area is dry sub-humid to moist sub-humid with average annual rainfall at 1470 mm, mostly falling during monsoon season from mid-June to September-October, often causing severe flood mainly in the upper reaches, more often two-three times in a year. Out of total 11.7%, only 43.7% of the net irrigated area. It means the agriculture of Bhagalpur depends heavily on rainfall. But, the pattern of rainfall was highly erratic in this area.

### **Geology & Hydrogeology**

Whole area has been divided into four different zones: Recent Alluvium (Non Calcareous), Recent Alluvium (Calcareous), Tal land soils and Old Alluvium. Gangetic plain has huge amount of river deposited sediments and accumulate at the flood and deltaic plain of Bengal. They generally consist of Pleistocene and Holocene formations. However central Gangetic plain (in eastern Bihar) have a unique geological-geomorphological setting with aquifer ranging from Quaternary Himalayan alluvial fans and plains to the pre-Cenozoic (Precambrian to Cretaceous) Indian cratonic, igneous and metasediment shield provinces. The east Ganga regions are mainly made up of flood plains and piedmont plains of Holocene. The coarse-grained detrital sediments dominate the channel fill deposits where as the over bank deposits comprise fine-grained clastic sediments in the Holocene alluvium. The alluvial central Gangetic plain slope from north to south, smooth on a regional scale but interrupted locally by sandy ridges and basins. The plain has been incised by dendritic drainage, with channels filled by organic mud of Holocene age. The geomorphology of the area is monotonously flat and featureless plain with gradient towards the river Ganges.

Most of the present wells used by the local communities as a whole are present in these aquifers, which are used extensively for drinking and agricultural purposes.

Recently Central Ground Water Board (CGWB) started putting wells below 300 m on large scale for community water supply. The groundwater occurs in Holocene sandy sediments and forms an extensive unconfined to leaky confined aquifers. i.e. the area has both confined and unconfined aquifer. Water level fluctuates with seasonal recharge and discharge. The depth of well varies from 6 m below ground level (bgl) to 13 m bgl. The depth to water level has been observed to vary from 3 to 8 m bgl during pre-monsoon and 2 to 4 m bgl during post-monsoon. The multiple aquifer system of this region has variable hydraulic conductivity and water quality. The deeper aquifers i.e. semi confined aquifers are prolific aquifers and may be a future water resource supply for the region with good hydraulic characteristics. This district has two ways for irrigation: (a) canals and (b) tube wells (groundwater).

### **HAZIPUR**

#### **G Location and Area**

Ghazipur district is located in the eastern alluvial part (classification based on agro-climatic zonation) of the Uttar Pradesh ranges from 25° 19' to 25° 54' N latitude and from 83° 4' and 83°58' E longitude (Map1). According to the central statistical organisation the district had an area of 3,381 sq. km. on 2001. The district of Ghazipur forms, the eastern part of the Varanasi Division. It lies to the east and north of the Jaunpur and Varanasi districts respectively between the parallels north latitude and east longitude. It is bounded on the north-west by Azamgarh, on the north-east by Ballia and on the south-east by the Shahabad district of Bihar, from which it is separated by the Karamnasa River.

#### **Demography**

As of 2001 India census, Ghazipur had a total population of 15,57,897 out of which rural portion occupies 14,62,654 and urban occupies 95,243. Males constitute 53% of the population and females 47 %. Ghazipur has an average literacy rate of 69 %, with 76 % of the males and 62% of the females literate.

#### **Geomorphology**

##### **River system**

The Ganga is the most important river of the district, the rivers Gomati, Gangi, Besu, Mangai, Sarju and the Karamnasa being directly or indirectly its main affluents. The Ganga - This river first touches this district in the extreme south-of pargaoa Saidpur and forms the boundary between Ghazipur and Varanasi for a long distance.

From Mainpur the Ganga sweeps to the north-east but a few kilometers beyond the district headquarters it bends to the south-east through a wide expanse of alluvium. At Bara the bed narrows and the stream flows eastward as far as Chaunsa in Shahabad, where it turns to the north-east separating pargana Muhammadabad from Bihar. The total length from its first point of contact in this district to its exit at Rasulpur is about 102 km.

Gomati - The first tributary to join the Ganga in this district which flows along the southern boundary of the Saidpur tahsil, separating it from Varanasi. The Gomati is here a navigable stream subject at times to heavy floods which are intensified by the action of the Ganga in holding up the water at the junction. The land along the Gomati is broken by ravines some of which extend inland for a long distance, one of the chief being the Sarwa, a small watercourse that forms the boundary between this district and Jaunpur.

The Gangi - The next tributary on the left bank is the Gangi, a stream rises near Jaunpur and makes its way in a south-easterly direction through the clay lands along the borders of Jaunpur and Azamgarh.

The Sarju - The Sarju (often known as the Tons and also as the Chhoti Sarju to distinguish it from the greater river of this name) contains the combined water of the Tons and the Chhoti Sarju, the two streams uniting near Mau in Azamgarh and then passing into pargana Zahurabad near Bahadurganj.

### **Climate and Rainfall**

The district has an extreme type of climate with hot summers and cold winters. The year may be divided into four seasons. The cold season from about the middle of November to February is followed by the summer season from March 10 about the middle of June. The period from mid-June to the end of September constitutes the south-west. Monsoon season, the succeeding period till the middle of November is the post-monsoon season. May is usually the hottest month with the mean daily maximum temperature at about 41°C and the mean daily minimum at about 26°C. The summer is intensely hot and on individual days the maximum temperature during May or early June occasionally exceeds 46°C. With the advent of the monsoon by about the middle of June there is an appreciable drop in the day temperature though the nights continue to be warm. Even in July and August, when the south-west monsoon is well established, day temperatures on some days during breaks in the rains may reach over 40°C. The day temperatures do not decrease even after the withdrawal of the monsoon early in October but the nights become cooler. After October both the day and night temperatures decrease rapidly. January is generally the coldest month with the mean daily maximum temperature at about 23°C and the mean

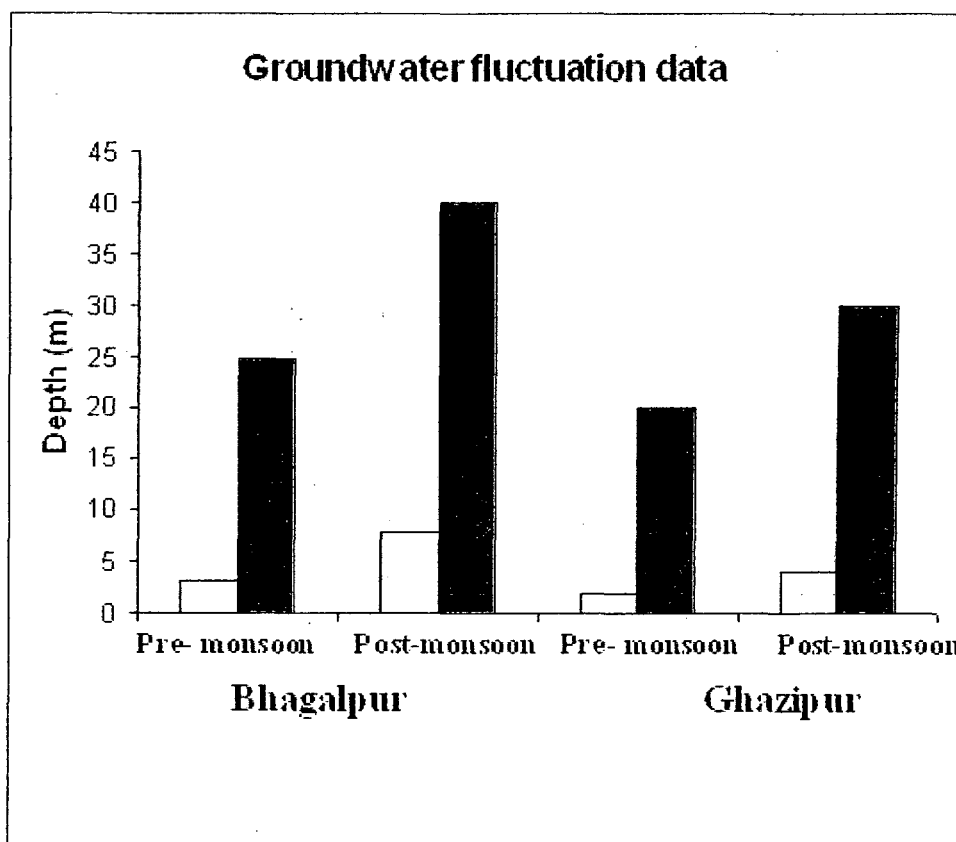
daily minimum at about 9°C. The south-west monsoon usually arrives in the district by about the middle of June and withdraws by the end of September. The average annual rainfall of the district is 1110 mm. During July and September, the relative humidity is high, being over 70 per cent. During the post-monsoon and winter seasons, the humidity is high in the morning. By summer, the relative humidities become very low, particularly in the afternoons from March to May when they are less than 25 per cent.

### **Geology & Hydrogeology**

The most valuable and most common mineral is kankar (limestone conglomerate) which occurs in extensive strata throughout the Upland area, at varying depths below the surface. Where possible it is quarried for ballast, road metal and for lime-burning. Clay suitable for making bricks is obtained everywhere. The district can be divided into three physical divisions the northern uplands; the central lowlands and the southern uplands. The northern uplands, which comprise all the country north of the Ganga, with the exception of parts of Saidpur and Ghazipur and the greater portion of Muhammadabad, mostly resemble the ordinary uplands of the Gangetic plain. The higher levels are sandy and on the slopes inland from the banks of the streams the soil becomes good fertile loam (usually of somewhat light character) but in the depressions this merges into a stiff clay which turns into usar wherever saturation occurs and the grey dhankur being fitted (as its name implies) for the cultivation of The light sandy soil is generally known as balua, the loam as doras (corresponding to dumat of other parts) and clay as matiyar. In most parts, of the upland there is a large amount of kankar in the subsoil. The alluvial lowlands, generally known as- terai comprise a small portion of the Saidpur and Ghazipur parganas, almost the whole of Karanda and the greater part of Muhammadabad from the Ganga to the road which goes from Ghazipur to Lathudih and Ballia. There are two karail (dark soil) tracts, one in Muhammadabad. The central portion of Zamania constitutes the southern uplands which have the ordinary loam and clay soils of the northern uplands. There is a stretch of fertile karail on the north of the Karamnasa river from the boundary of Varanasi on the west to the village of Dewalon the east.

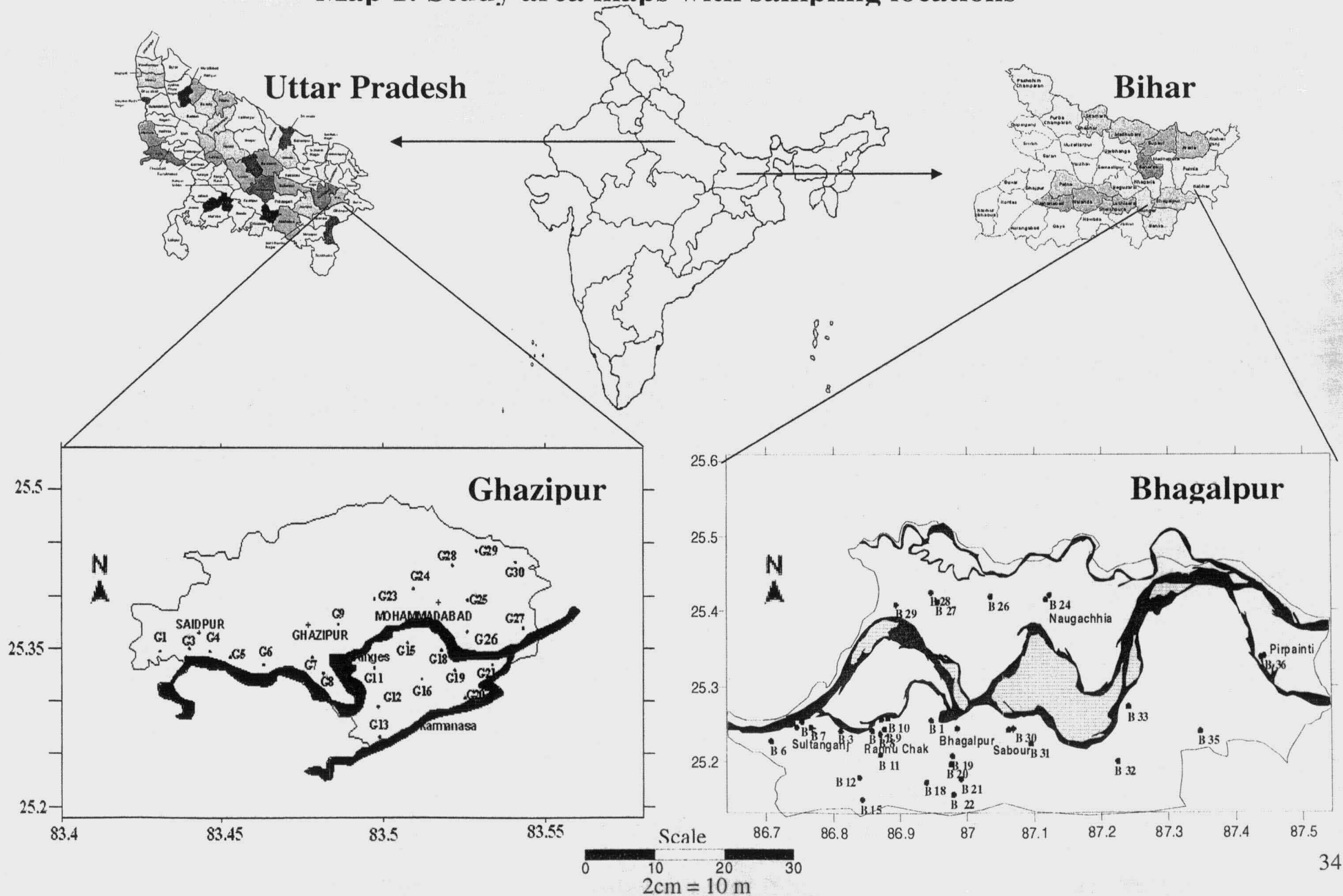
Most of the present wells used by the local communities as a whole are present in these aquifers, which are used extensively for drinking and agricultural purposes. Recently Central Ground Water Board (CGWB) started putting wells below 150 m on large scale for community water supply. The groundwater occurs in Holocene sandy sediments and forms an extensive unconfined to leaky confined aquifers. i.e. the area has both confined and unconfined aquifer. Water level fluctuates with seasonal recharge and discharge. The depth of well varies from 20 m bgl to 40 m

bgl. The depth to water level has been observed to vary from 25 to 40 m bgl during pre-monsoon and 20 to 30 m bgl during post-monsoon. The multiple aquifer system of this region has variable hydraulic conductivity and water quality. The deeper aquifers i.e. semi confined aquifers are prolific aquifers and may be a future water resource supply for the region with good hydraulic characteristics. This district has two ways for irrigation: (a) canals and (b) tube wells (groundwater).



**Graph No.- A.** Source- District Information Centre (Bhagalpur, 2006 and Ghazipur, 2005-06)

Map 1: Study area maps with sampling locations





## **F**ield Methods

Initially, to understand the general variation in groundwater chemistry over the study area, a well inventory survey was carried out during May 2007 and October 2007 and electrical conductivity (EC) and pH were measured. A global positioning system (GPS) was used for location and elevation reading (Topographic sheets made available from Geological Survey of India supported this). These data were used to select the representative wells and hand pumps for groundwater sampling. Sampling wells were selected in such a way that they represent different geological formations as well as land-use pattern at varying topography of this area. A total 132 ground water samples were collected in the flood plain of central Gangetic plain of District Bhagalpur (Bihar) and Ghazipur (Uttar Pradesh) during May 2007 to October 2007 in both the season of pre and post-monsoon from shallow and deep aquifers. Latitude and longitude of all the sampling locations are mentioned in appendix (Table i & ii).

Water samples were collected in clean polyethylene bottles. At the time of sampling, bottles were thoroughly rinsed 2-3 times with groundwater to be sampled. In the case of bore wells and hand pumps, the water samples were collected after pumping the water for 10 minutes. In case of open wells, water samples were collected 30 cm below the water level using a depth sampler. This was done to remove groundwater stored in the well itself and to obtain representative samples. In-situ measurements included EC, pH, ORP, Arsenic and bicarbonate that were measured using a portable Orion Thermo water analyzing kit (Model Beverly, MA, 01915) and titration respectively as per WHO (1993) recommendation that this should be made in-situ, or in the field immediately after water sample has been obtained, since these parameters change with storage time. Total Arsenic was tested with the help of Digital Arsenator (Wagtech Company, UK) and all the values were cross checked by Atomic Absorption Spectrophotometer (Shimadzu AA-6800) while Arsenic speciation was performed with Disposable Cartridges (MetalSoft Center, PA) in the field which absorbs As(V), but allow As(III) to pass through. Further, these samples were stored below 4°C in portable ice-box to minimize the chemical alterations. The collected groundwater samples were classified for anions and cation analysis. Further, groundwater samples were filtered by 0.45 µm Millipore filter paper and acidified with 2N HNO<sub>3</sub> (Ultra pure Merck) for cation analysis and HBO<sub>3</sub> acid

was used as preservative for nitrate analysis. Fe and Mn were analyzed in the laboratory by Atomic Absorption Spectrophotometer (Shimadzu AA-6800). Concentrations of total arsenic were cross checked on acidified samples using graphite-furnace (GF) AAS (Shimadzu AA-6800) on wavelength in absorption mode using chemicals standards with detection limit of 2 µg/l. Major cation analysis ( $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) was carried out by EEL Flame Photometer (APHA, 1995). Main anions  $\text{F}^-$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$  were analysed by the Dionex Ion-Chromatography DX-120. Other parameters like  $\text{SiO}_2$  and  $\text{NH}_4^+$  was analysed with JENWAY Spectrophotometer model 6505. High purity reagents (Merck) and milli-Q water (Model Milli-Q, Biocel) were used for all the analysis.

#### **pH and Conductivity (EC)-**

The pH and conductivity was measured in unfiltered water samples. CD-MOS used technique pH meter was used to measure the pH which makes the instrument more versatile and reliable. The pH was measured by Orion Thermo water analyzing kit (Model Beverly, MA, 01915). The electrode was conditioned (0.1 M KCl solution) and calibrated with buffer solutions of pH 4, 7 and 9.2. The temperature knob was set in accordance with the temperature of the water samples. The samples were stirred continuously in order to maintain homogeneity before noting down the pH.

The conductivity was measured in µS/cm using Orion Thermo water analyzing kit (Model Beverly, MA, 01915) Conductivity Meter. The instrument was calibrated and set for 0.01M KCl standard (1413 µS/cm at 25°C).

#### **ORP-**

This parameter has been carried out by using same instrument i.e. ORP electrode was used for this parameter (Platinum electrodes were used for ORP)

#### **Bicarbonate-**

The bicarbonate content was determined by the potentiometer titration method (APHA-1995). Bicarbonate standards ranging from 100-1000 mg/l<sup>-1</sup> was prepared from  $\text{NaHCO}_3$ . 50 ml sample and a series of bicarbonate standards were titrated against 0.02N HCl. The end point was noted at pH 4.5. A standard graph was plotted between bicarbonate standards and volume of acid consume. The readings of samples were found out from this graph.

**Laboratory methods-****Major Cations**

- **Sodium, Potassium and Magnesium**

These were analyzed using AIMIL Flame photometer.

- **Calcium**

The analysis of major cation was carried out on Shimadzu-AA-6800, Atomic absorption Spectrophotometer. Calcium was analyzed in absorption mode at the wavelength of 422.7nm. The instrument was calibrated with different chemical standard.

**Heavy metal- Iron and Arsenic**

Fe and Mn metal analyses were done by using Shimadzu-AA-6800, Atomic absorption Spectrophotometer (AAS).

For Arsenic detection Grafite-furnace-AAS was used. First the instrument Atomic absorption Spectrophotometer (AAS) was calibrated by using known concentration of arsenic sample and a standard curve was drawn between absorbance and concentration, from this graph concentration of filtered water samples were calculated.

**Contour Diagram**

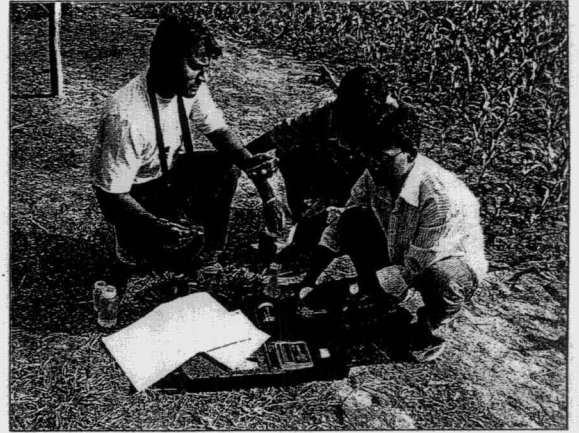
Contour Diagram of various water quality parameter concentrations were drawn by using Golden Surfur (8) software. After geo-referencing by using Latitude and Longitude values of sampling point's contours are drawn in SURFUR ver.8.

**Water type Diagram**

Piper, Schoeller, Durov and different scattered diagrams were plotted using Aq.QA Software to understand water type and their characteristics.

**Statistical Analysis**

With help of "Statistical Package for Social Sciences (SPSS), version-10.0". software different statistical analysis (factor analysis and Correlation matrix) have been done in order to well interpretation of data. To evaluate the potential relationship between various physicochemical parameters, Pearson correlation was carried out. Factor analysis for groundwater samples was carried out by using "Principal component analysis" and "Varimax Rotation" was used for extraction and deriving factors respectively.



Analysis of Arsenic and physical parameters on field with colleagues



Carrying out public awareness campaign at field about the precautions to be followed before water consumption

## ***G*** eochemistry of Groundwater of Bhagalpur, Bihar

The analytical precision for the measurement of ions was determined by calculating the Normalized inorganic Charge Balance (Huh et al 1998), which is defined as  $[Tz^+ - Tz^- / Tz^+ + Tz^-]$  and represents the fractional difference between the total cations and total anions (Edmond et al 1995). As exemplified by (Huh et al 1998) the measured major ions ( $Na^+$ ,  $K^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Cl^-$ ,  $SO_4^{2-}$ ,  $HCO_3^-$ ,  $NO_3^-$ ) are generally enough to give a charge balance. Most of the ground water samples showed a charge imbalance mainly in favour of positive charge. The observed average charge balance between cations ( $Tz^+$ ) and anions ( $Tz^-$ ) support the quality of the data points, which is  $\pm 7.92\%$  (average) in Pre-monsoon where as it is  $9.12\%$  (average) in Post-monsoon. This imbalance of  $\Delta Tz$  could be related to the fact that no analysis was made on organic matter, which is mainly produced by biological activities during the Pre-monsoon and Post-monsoon (Berner-Kay and Berner 1987; Edmond et al. 1995; Huh et al. 1998).

The ground water was alkaline as evident from the average value of pH, which was 8.1 and 8.0 in the Pre-monsoon and Post-monsoon respectively (Table 7 & 8). The precipitation induced dissolution processes might have affected the pH of ground water. The high pH in the Pre-monsoon suggests that the soils/sediments are very reactive, which eventually enhances dissolution (Subramanian and Saxena 1983).

The value of EC varied from 250 to 980  $\mu S/cm$  in the Pre-monsoon and 217 to 968  $\mu S/cm$  in the Post-monsoon (Table 7 & 8). The higher value of EC in the Pre-monsoon suggests the high concentration of dissolved solids and/or high ionic strength of groundwater, local variation in soil type, multiple aquifer system and agricultural activities in the area.

Bicarbonate represents the major source of alkalinity. In Bhagalpur, bicarbonate concentration ranged between 15.2 to 214.5 mg/L in the Pre-monsoon and 23.7 to 266.3 mg/L in the Post-monsoon (Table 7 & 8). High  $HCO_3^-$  concentration in the Post-monsoon period indicates the weathering of carbonaceous sandstones in the aquifer, while low concentration in the Pre-monsoon period may be due to the precipitation of  $HCO_3^-$  along with other cations.

Parameters	Unit	Minimum	Maximum	Average
Depth	(ft)	30	95	55.67
Ph		7.8	8.3	8.13
ORP	mv	-134	169	8.97
EC	µs/cm	250	980	599.17
TDS	mg/L	191.25	710	455.22
Na <sup>+</sup>	mg/L	5.76	41.91	18.66
K <sup>+</sup>	mg/L	0.44	3.22	1.45
Ca <sup>+2</sup>	mg/L	20.7	126.03	64.81
Mg <sup>+2</sup>	mg/L	7.83	18.2	10.67
HCO <sub>3</sub> <sup>-</sup>	mg/L	15.2	214.5	104.85
F	mg/L	0.08	4.94	0.76
Cl	mg/L	6.56	219.93	69.67
NO <sub>3</sub> <sup>-</sup>	mg/L	1.01	39	23.71
SO <sub>4</sub> <sup>2-</sup>	mg/L	3.84	72.8	32.93
PO <sub>4</sub> <sup>3-</sup>	mg/L	2.66	6.37	4.05
SiO <sub>2</sub>	mg/L	14	49.33	30.01
NH <sub>4</sub> <sup>+</sup>	mg/L	0.59	3.11	1.38
Fe	mg/L	0.66	7.62	3.19
Mn	mg/L	0.01	1.79	0.66
As(tot)	µg/L	19.1	118	51.23
As <sup>3+</sup>	µg/L	10.9	81.1	34.43
As <sup>5+</sup>	µg/L	7.1	55.2	16.81

Table 7 Statistical analysis of Pre-monsoon data of Bhagalpur (n=36)

Parameters	Unit	Minimum	Maximum	Average
Depth	(ft)	30	95	56.03
Ph		7.66	8.16	7.98
ORP	mv	-125.41	135	7.91
EC	µs/cm	216.85	967.64	591.13
TDS	mg/L	184.72	685.74	441.4
Na <sup>+</sup>	mg/L	5.16	37.58	17
K <sup>+</sup>	mg/L	0.43	3.16	1.39
Ca <sup>2+</sup>	mg/L	18.87	160.92	69.4
Mg <sup>2+</sup>	mg/L	8.36	19.43	11.5
HCO <sub>3</sub> <sup>-</sup>	mg/L	23.74	266.34	129.42
F	mg/L	0.06	2.48	0.63
Cl	mg/L	6.08	154.72	55.52
NO <sub>3</sub> <sup>-</sup>	mg/L	1.12	46.7	22.66
SO <sub>4</sub> <sup>2-</sup>	mg/L	3.61	68.43	27.68
PO <sub>4</sub> <sup>3-</sup>	mg/L	2.36	5.65	3.59
SiO <sub>2</sub>	mg/L	17.27	55.55	35.1
NH <sub>4</sub> <sup>+</sup>	mg/L	0.54	2.83	1.26
Fe	mg/L	0.43	6.84	2.82
Mn	mg/L	0.01	1.75	0.63
As(tot)	µg/L	18.46	113.49	48.97
As(3+)	µg/L	11.63	75.9	32.73
As(5+)	µg/L	5.32	47.89	16.24

Table 8 Statistical analysis of Post-monsoon data of Bhagalpur (n=36)

The  $\text{SO}_4^{2-}$  concentration varied from 3.84 to 72.8 mg/L in Pre-monsoon and 3.61 to 68.43 mg/L in Post-monsoon (Table 7&8). This indicates that the sulfate enrichment is high in Pre-monsoon due to breakdown of organic material and agriculture runoff carrying unutilized  $\text{SO}_4^{2-}$  (Anderson et al 1979). The groundwater may have secondary salinity, as indicated by high  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  concentrations.

In this study,  $\text{Cl}^-$  varied from 6.56 to 219.9 mg/L in the Pre-monsoon and 6.08 to 154.72 mg/L in the Post-monsoon (Table 7 & 8). The higher and lower concentration of  $\text{Cl}^-$  in the Pre-monsoon and post monsoon may be due to the input from sewage effluents in the village areas and dilution by rain water in post monsoon season (Todd 1980). The physical processes such as mixing with another aquifer of different  $\text{Cl}^-$  concentrations and change in evaporation rate during recharge either spatially or temporally may cause the seasonal changes in the chloride content of groundwater. The high concentrations of  $\text{Cl}^-$  suggests mixing of fresh groundwater with soil water rich in dissolved ions derived from the salt precipitate coating on soils after Pre-monsoon season i.e. may be leaching during monsoon season. The presence of salts in unsaturated top zone of groundwater suggests that the flushing rate of the aquifers may be slow due to high  $\text{Cl}^-$  concentration trapped in clayey lenses, which may be gradually diffusing into the aquifer.

In Bhagalpur,  $\text{NO}_3^-$  varied significantly from 1.01 to 39 mg/L in Pre-monsoon and 1.12 to 46.70 mg/L in Post-monsoon (Table 7 & 8), which indicates the contact of groundwater to agricultural activities and microbial mineralization. Distribution pattern of it for Pre-monsoon and Post-monsoon has been shown in fig. (8 e&f) respectively. The higher concentration of it is caused by the presence of *E. coli*, *Staphylococcus aureus*, *Proteus vulgaris*, *Salmonella typhi* and *Pseudomonas aeruginosa* indicated the faecal contamination reported by (Saha et al. 2006).

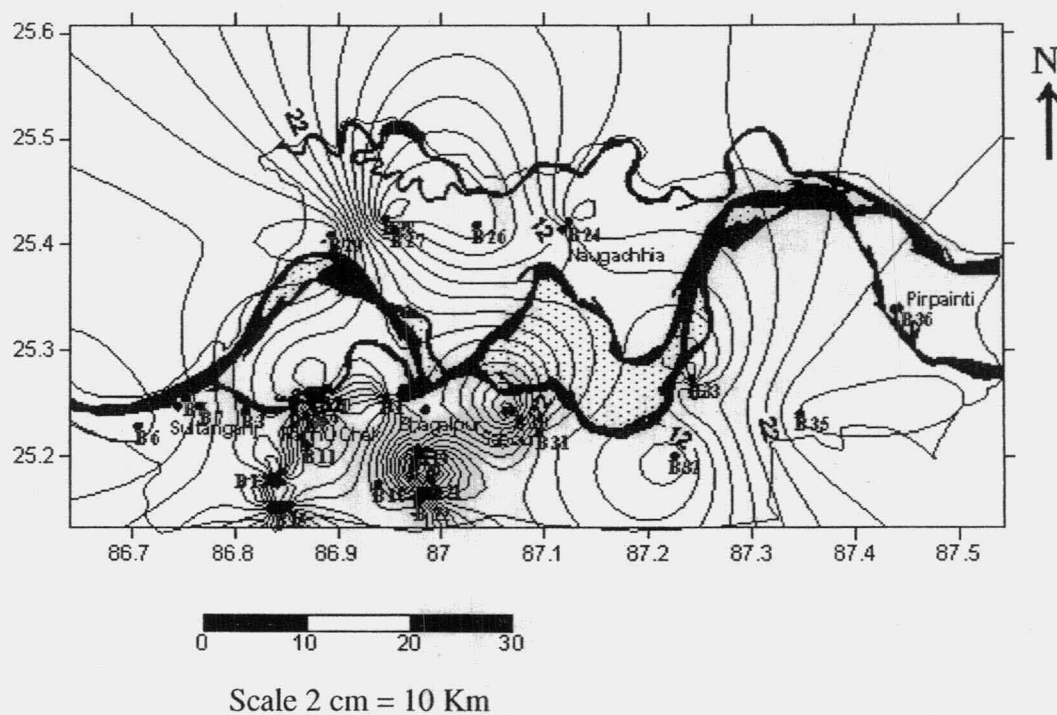


Fig.-8 (e) Contour map of nitrate for pre monsoon in Bhagalpur

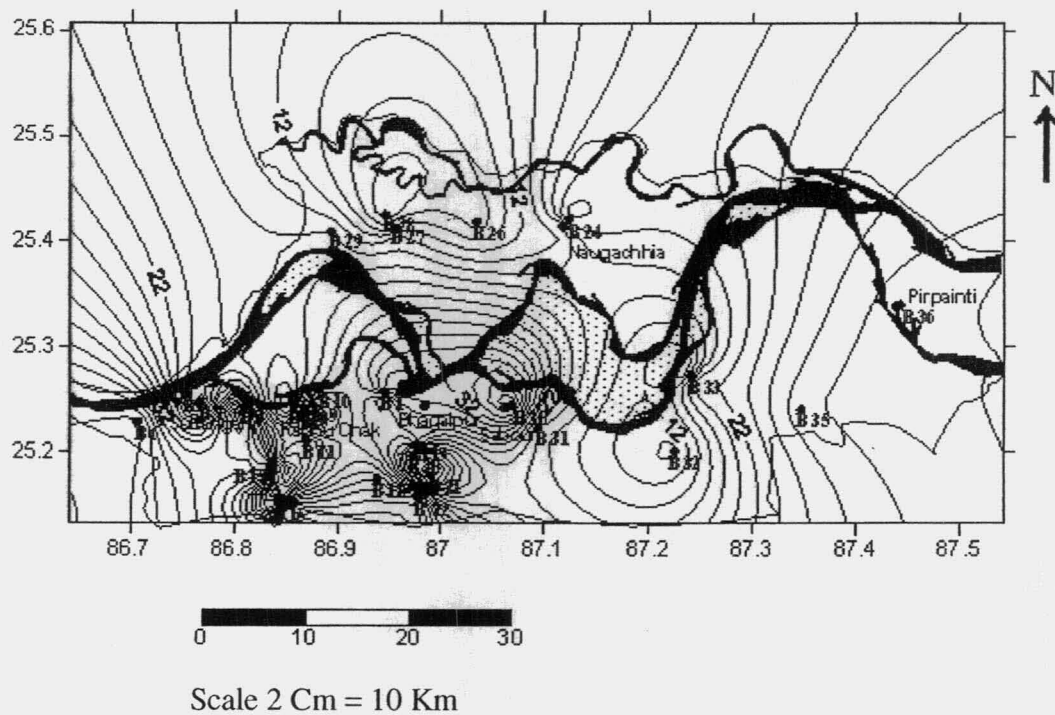


Fig.-8 (f) Contour map of nitrate for Post- monsoon in Bhagalpur



The concentration of  $\text{PO}_4^{3-}$  varied from 2.66 to 6.37 mg/L in the Pre-monsoon while in the Post-monsoon it ranged between 2.36 to 5.65 mg/L (Table 1 & 2). The underlying cause of  $\text{PO}_4^{3-}$  in groundwater of Bhagalpur district indicates input of fertilizers in farmlands to enhance the paddy and wheat productivity and dilution effect in rainy season.

In Bhagalpur,  $\text{F}^-$  varied significantly from 0.08 to 4.94 mg/L in Pre-monsoon and 0.06 to 2.48 mg/L in Post-monsoon (Table 7 & 8), which is indicator of weathering of mica containing mineral like apatite. Its pollution and health aspect was also reported earlier by Chaurasia et al. (2007).

Finally among anions bicarbonate is the dominant species while average value trend found was  $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^- > \text{PO}_4^{3-} > \text{F}^-$  in both period.

The dominant cation was  $\text{Ca}^{2+}$  followed by  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$  and  $\text{NH}_4^+$ . There is a slight variation in seasonal distribution of all the ions and are significant at certain locations. The concentration of  $\text{Ca}^{2+}$  varied from 20.7 to 126 mg/L in Pre-monsoon and 22.63 to 160.92 mg/L in Post-monsoon season (Table 7 & 8). In Post-monsoon season high concentration of  $\text{Ca}^{2+}$  may be due to weathering of carbonate mainly from gypsum and plagioclase feldspar minerals, which is abundant in flood plain regions (Bhattacharya et al. 1997). In the Pre-monsoon the  $\text{Mg}^{2+}$  concentration varied from 7.83 to 18.2 mg/L while in the Post-monsoon it was ranged between 8.36 to 19.43 mg/L (Table 7 & 8).

However the results for  $\text{Na}^+$  and  $\text{K}^+$  followed the opposite pattern with high concentrations in the Pre-monsoon (Table 7 & 8). This infers that the contribution of cations via alumino-silicate weathering is low in comparison to carbonate weathering. The average ratio trend of  $(\text{Ca}^{2+} + \text{Mg}^{2+}) / (\text{Na}^+ + \text{K}^+)$  varied from 3.75 in the Pre-monsoon and 4.41 in the Post-monsoon indicates the dominance of carbonate rock weathering in the Bhagalpur ground water.

The concentration of  $\text{NH}_4^+$  varied from 0.59 to 3.11 mg/L in Pre-monsoon and 0.54 to 2.83 mg/L in Post-monsoon season (Table 7 & 8). This higher concentration indicates a possible contamination through the reduction of nitrate rich mineral by microbes (Bhattacharya et al. 2007).

Table 1 Hydro geochemistry of samples in Pre-monsoon of Bhagalpur (n=36)

ID	Depth (ft)	Type*	Ph	ORP mv	EC µs/cm	TDS mg/L	Na <sup>+</sup> mg/L	K <sup>+</sup> mg/L	Ca <sup>2+</sup> mg/L	Mg <sup>2+</sup> mg/L	HCO <sub>3</sub> <sup>-</sup> mg/L	F <sup>-</sup> mg/L	Cl <sup>-</sup> Mg/L	NO <sub>3</sub> <sup>-</sup> mg/L	SO <sub>4</sub> <sup>2-</sup> mg/L	PO <sub>4</sub> <sup>3-</sup> mg/L	SiO <sub>2</sub> mg/L	NH <sub>4</sub> <sup>+</sup> mg/L	Fe mg/L	Mn mg/L	As(tot) µg/L	As <sup>3+</sup> µg/L	As <sup>5+</sup> µg/L
B 1	30	HP	8.2	-14	430	329	14.28	1.10	31.04	8.05	103.7	0.56	13.47	19.94	8.5	4.32	18.41	1.2	5.45	0.02	63	43	20
B 2	50	TW	8.2	-54	520	398	15.20	1.17	73.59	10.64	88.3	0.41	75.78	35.9	39.72	4.19	23.25	1.1	4.16	0.27	70	31.6	38.4
B 3	45	HP	8.2	-65	610	467	20.49	1.57	73.68	10.56	109.8	0.46	94.46	25.02	26.82	3.98	26.90	2.45	4.31	0.01	88	32.8	55.2
B 4	42	HP	8.1	-66	810	620	14.74	1.13	81.1	11.13	130.5	0.69	113.8	25.63	28.74	5.4	28.02	1.57	5.61	0.13	71	52.8	18.2
B 5	51	HP	8.2	-65	540	387	24.41	1.87	57.82	9.36	81	1.52	54.46	38.67	19.63	4.17	21.10	1.31	6.3	0.27	87	67	20
B 6	37	HP	7.9	-71	590	410	12.97	0.92	40.21	10.9	115.4	0.5	58	32.63	15.01	3.65	34.90	1.44	5.89	0.01	65	51	14
B 7	48	HP	8.1	-87	310	231	22.80	1.75	26.42	8.96	36.6	0.36	54.01	29.82	18.43	5.21	17.19	1.09	5.12	0.03	54	36.3	17.7
B 8	55	HP	8.1	-106	540	413	18.70	1.44	63.5	14.4	88.3	0.45	56	36.2	36	5.85	37.68	1.55	4.7	0.09	77	57.1	19.9
B 9	50	HP	8	-112	630	482	23.00	1.77	106	17.2	98.8	0.52	117	39	31.27	4.29	33.73	1.24	4.2	0.12	59	43.8	15.2
B 10	35	HP	8.2	-134	570	436	19.50	1.50	76	18.2	93.1	0.67	103	18	51.8	6.37	28.28	2.28	6.76	0.04	118	81.1	36.9
B 11	55	HP	8.1	78	410	314	7.60	0.58	61.4	11.07	34.2	0.65	54.72	38.1	22.81	3.13	36.15	0.95	2.26	0.93	33.7	18.2	15.5
B 12	65	HP	8.2	90	370	283	11.97	0.92	46.63	11.15	62.2	1.03	36.94	15.8	27.56	3.11	20.47	0.88	1.62	1.02	25.8	12.1	13.7
B 13	60	TW	8.1	78	550	421	16.20	1.24	57.54	10.3	121.1	0.67	33	37.2	26.2	3.05	32.90	0.92	1.12	1.09	27.43	16.21	11.22
B 14	63	OW	8.2	169	590	451	17.33	1.33	51.12	8.69	143.5	0.51	71	36.4	31.98	2.95	33.14	0.79	1.34	1.47	24.3	10.9	13.4
B 15	72	HP	7.8	86	250	191	10.13	0.78	20.7	10.04	15.2	0.14	52.72	15.02	14.12	2.83	16.56	0.71	1.3	1.2	31.5	22.2	9.3
B 16	57	OW	8.2	115	790	604	23.26	1.78	106.4	10.36	109.8	0.53	95.54	38.82	69.48	2.66	45.02	0.65	1.39	0.92	22.6	12.9	9.7
B 17	45	HP	8.3	97	420	321	12.89	0.99	41.74	10.8	99.8	0.85	33.66	24.18	19.91	3.46	49.31	0.81	1.56	1.1	31.4	21.7	9.7
B 18	66	HP	8.2	94	510	390	13.59	1.04	46.39	10.9	61	0.81	66.37	28.31	22.88	4.01	38.07	0.98	1.68	0.95	28.8	18.6	10.2
B 19	50	HP	7.9	86	980	710	29.01	2.23	124.1	7.83	170.8	0.4	123.6	22.2	70.84	3.34	40.76	0.64	1.19	1.12	19.1	11.5	7.6
B 20	75	HP	8.1	112	670	513	38.91	2.99	29.09	10.73	214.5	2.55	14.15	3.9	11.52	3.56	32.55	0.73	0.87	1.38	28.2	18.8	9.4
B 21	70	OW	8.2	119	710	543	29.14	3.01	30.21	9.21	174.5	4.94	82.6	1.01	49.28	4.06	29.21	0.59	0.91	1.31	22.4	15.3	7.1
B 22	75	HP	8	130	840	643	41.91	3.22	101.2	9.32	72.2	1.34	193	30.2	63.92	4.23	37.67	0.92	0.83	1.31	38.7	26.6	12.1
B 23	80	TW	7.8	101	910	696	26.02	2.00	98.97	7.83	131	0.59	129.1	32.67	50.95	3.38	40.17	1.12	1.74	0.94	33.6	24.4	9.2
B 24	40	OW	8.1	-120	510	390	31.55	2.42	48.65	10.71	103.7	0.3	42.63	21.2	23.25	4.31	35.16	2.54	4.54	0.09	64	56.6	7.4
B 25	35	HP	8.2	-102	540	413	10.13	0.78	44.07	10.76	61	0.21	54.78	12.31	29.8	4.77	30.59	1.63	4.23	0.23	52.6	41.1	11.5
B 26	45	HP	8.2	-103	490	375	9.21	0.71	40.78	10.83	125.9	0.45	10.92	6.23	12.15	4.68	23.15	2.55	5.87	0.25	79	57.7	21.3
B 27	30	HP	8.3	-54	610	467	8.75	0.67	57.76	10.42	61	0.3	60.4	12.6	23.23	4.57	21.96	2.83	4.17	0.24	66	46.1	19.9
B 28	30	HP	8.3	-106	370	283	5.76	0.44	33.06	8.08	92.5	0.19	9.74	3.3	3.84	5.67	14.00	3.11	7.62	0.03	117.4	79.9	37.5
B 29	35	HP	8.2	-131	550	421	10.13	0.78	85.86	14.11	122	0.42	34.41	32.82	47.67	5.22	36.25	2.99	6.06	0.11	71.6	49.8	21.8
B 30	38	HP	8.2	-58	860	658	22.34	1.71	125.1	10.47	61	0.65	166.8	36.55	72.71	5.41	25.09	2.78	6.44	0.08	94	75.6	18.4
B 31	45	HP	8.1	52	760	581	12.89	0.99	57.4	9.78	143.5	0.31	12.5	15.67	10.45	3.35	24.37	1.04	0.88	0.78	27.3	14.1	13.2
B 32	95	HP	8.3	48	510	390	11.74	0.90	41.66	11.05	126.9	0.42	6.56	8.2	13.62	3.77	26.78	0.94	0.93	0.8	37.7	24.3	13.4
B 33	80	HP	8.2	67	720	551	22.57	1.73	110	10.35	164.7	0.08	68.72	8.24	58.51	3.29	30.38	1.01	0.98	0.89	31.5	20.7	10.8
B 34	85	HP	8.2	74	650	497	24.41	1.87	66.55	9.53	91.5	1.45	69.11	20.26	45.72	3.16	33.31	0.91	1.08	0.98	32.3	19.9	12.4
B 35	80	TW	8.1	72	540	413	14.51	1.11	51.46	10.44	128.1	0.81	25.4	26.68	14.2	3.28	28.62	0.82	1.18	1.79	30.9	16.3	14.6
B 36	90	HP	8.1	103	910	696	23.72	1.82	126	10.12	137.4	0.47	219.9	25.03	72.8	3.05	29.41	0.71	0.66	1.76	20.6	11.4	9.2

(HP-Hand Pump, OW- Open Well, TW- Tube Well)

Table 2 Hydro geochemistry of samples in Post-monsoon of Bhagalpur (n=36)

ID	Depth (ft)	Type	Ph	ORP mV	EC µs/cm	TDS mg/L	Na <sup>+</sup> mg/L	K <sup>+</sup> mg/L	Ca <sup>2+</sup> mg/L	Mg <sup>2+</sup> mg/L	HCO <sub>3</sub> <sup>-</sup> mg/L	F <sup>-</sup> mg/L	Cl <sup>-</sup> mg/L	NO <sub>3</sub> <sup>-</sup> mg/L	SO <sub>4</sub> <sup>2-</sup> mg/L	PO <sub>4</sub> <sup>3-</sup> mg/L	SiO <sub>2</sub> mg/L	NH <sub>4</sub> <sup>+</sup> mg/L	Fe mg/L	Mn mg/L	As(tot) µg/L	As <sup>3+</sup> µg/L	As <sup>5+</sup> µg/L
B 1	30	HP	8.1	-13	425	317.7	12.80	0.95	32.50	11.20	114.5	0.54	11.40	20.20	7.80	3.83	22.72	1.09	4.61	0.02	58.5	45.9	12.6
B 2	50	TW	7.9	-51	513	384.2	13.63	1.02	54.50	11.36	23.7	0.36	68.30	34.16	37.38	3.72	28.89	0.98	3.73	0.22	67.7	33.7	33.9
B 3	45	HP	8.0	-61	602	450.7	18.38	1.47	86.27	11.27	142.4	0.42	87.60	27.74	25.24	3.53	33.19	2.23	3.75	0.01	82.9	35.0	47.9
B 4	42	HP	8.0	-58	778	598.5	13.21	1.11	137.20	11.10	169.3	0.61	105.52	46.70	27.05	4.79	34.57	1.43	5.03	0.13	65.6	56.4	9.2
B 5	51	HP	8.1	-54	512	373.8	21.89	1.84	47.10	9.99	105.1	1.48	50.50	22.88	18.47	3.70	33.44	1.19	4.80	0.26	84.1	41.4	42.7
B 6	37	HP	7.8	-66	685	487.0	10.74	0.90	64.38	11.63	110.8	0.49	66.80	43.30	14.13	3.24	18.38	1.31	5.28	0.01	62.8	54.4	8.4
B 7	48	HP	8.0	-80	306	223.1	20.44	1.72	23.99	9.40	47.5	0.30	27.60	13.06	17.34	4.62	27.12	0.91	4.59	0.03	52.2	38.7	13.5
B 8	55	HP	8.1	-99	533	399.0	16.77	1.41	63.17	15.37	114.5	0.42	49.80	40.14	33.88	5.19	46.49	1.41	4.22	0.09	74.4	60.9	13.5
B 9	50	HP	7.9	-99	621	465.5	20.62	1.73	103.12	18.36	128.1	0.49	72.30	36.51	47.10	3.80	24.62	1.13	4.10	0.12	57.0	46.7	10.3
B 10	35	HP	8.1	-125	563	421.2	17.49	1.47	66.39	19.43	115.6	0.53	81.80	23.22	18.75	5.65	34.89	2.08	5.93	0.02	101.7	66.5	35.2
B 11	55	HP	8.0	73	407	309.5	6.81	0.57	39.02	11.82	44.4	0.53	50.75	20.33	21.47	2.78	44.60	0.87	2.03	0.87	32.6	19.4	13.2
B 12	65	HP	8.1	64	361	273.4	10.74	0.85	27.73	11.90	35.8	0.92	44.26	9.69	25.94	2.76	29.93	0.80	1.45	1.00	24.9	12.9	12.0
B 13	60	TW	8.0	73	543	406.4	14.53	1.22	74.04	10.99	157.1	0.65	30.60	41.25	24.66	2.70	40.59	0.84	1.00	1.02	26.5	17.3	9.2
B 14	63	OW	8.1	135	580	435.9	15.54	1.25	65.78	9.28	146.1	0.43	65.84	15.60	30.10	2.62	40.89	0.72	1.20	1.44	23.5	11.6	11.9
B 15	72	HP	7.7	80	217	184.7	9.09	0.76	22.63	10.72	50.9	0.11	27.44	2.61	13.29	2.51	33.62	0.65	1.17	1.17	30.5	23.7	6.8
B 16	57	OW	8.1	108	780	583.7	20.85	1.66	125.80	11.06	142.4	0.52	88.60	43.04	65.39	2.36	55.55	0.59	1.25	0.90	21.8	13.8	8.1
B 17	45	HP	8.2	91	415	310.3	11.56	0.97	43.40	11.00	129.4	0.79	31.22	6.81	18.74	3.07	40.86	0.74	1.40	1.08	30.4	23.2	7.2
B 18	66	HP	8.1	88	504	376.8	12.18	1.02	79.87	11.63	79.1	0.79	61.55	31.39	21.53	3.56	46.97	0.89	1.51	0.91	27.8	19.9	8.0
B 19	50	HP	7.8	80	968	685.7	26.02	2.01	159.64	8.36	221.5	0.39	114.60	24.62	55.80	2.96	50.29	0.58	1.07	1.10	18.5	12.3	6.2
B 20	75	HP	8.0	105	662	495.0	34.89	2.73	37.43	11.45	256.0	2.48	13.12	4.32	10.84	3.16	40.16	0.66	0.78	1.35	27.3	20.1	7.2
B 21	70	OW	8.1	111	697	524.6	35.10	2.80	28.87	9.83	266.3	1.80	46.60	1.12	26.38	3.60	36.05	0.54	0.82	1.28	21.7	16.3	5.3
B 22	75	HP	7.9	122	829	620.6	37.58	3.16	130.27	9.95	85.8	1.30	118.94	33.49	60.15	3.75	46.48	0.84	0.66	1.23	37.4	18.7	18.7
B 23	80	TW	7.7	95	899	672.4	23.33	1.96	127.35	8.36	169.9	0.57	119.71	36.22	47.95	3.00	51.19	1.02	1.56	0.76	32.5	26.0	6.4
B 24	40	OW	8.0	-112	504	376.8	28.29	2.38	64.07	11.85	134.5	0.29	39.53	23.51	21.88	3.82	24.39	2.31	4.07	0.09	61.9	40.1	21.8
B 25	35	HP	8.1	-95	533	386.0	9.09	0.76	70.34	11.48	79.1	0.21	50.80	13.65	28.04	4.23	37.75	1.48	3.80	0.22	50.9	32.2	18.7
B 26	45	HP	8.1	-98	484	355.2	8.26	0.69	44.53	10.85	150.3	0.44	10.13	6.91	11.43	4.15	28.56	2.32	5.27	0.24	76.4	55.2	21.2
B 27	30	HP	8.2	-51	602	450.7	7.85	0.66	71.11	11.12	79.1	0.29	56.01	13.97	21.86	4.05	27.10	2.58	3.74	0.17	63.8	49.2	14.6
B 28	30	HP	8.2	-99	365	273.4	5.16	0.43	31.21	11.83	120.0	0.18	9.03	3.66	3.61	5.03	17.27	2.83	6.84	0.03	113.5	75.9	37.6
B 29	35	HP	8.1	-113	543	406.4	9.09	0.76	70.48	15.06	158.2	0.41	31.91	16.39	44.86	4.63	22.73	2.72	5.44	0.11	69.2	33.6	35.6
B 30	38	HP	8.1	-54	849	643.8	20.03	1.68	160.92	11.18	79.1	0.55	154.72	40.53	68.43	4.80	30.95	2.53	5.78	0.08	90.9	63.4	27.5
B 31	45	HP	8.0	51	746	561.5	11.56	0.97	53.93	10.44	186.1	0.30	11.59	17.37	9.83	2.97	30.07	0.95	0.79	0.76	26.4	15.0	11.3
B 32	95	HP	8.2	47	504	376.8	11.00	0.89	52.39	11.79	164.6	0.41	6.08	9.09	12.82	3.34	33.04	0.86	0.83	0.77	36.4	25.9	10.5
B 33	80	HP	8.1	63	701	521.0	20.23	1.70	73.51	11.05	213.6	0.06	63.73	9.14	16.20	2.92	37.48	0.92	0.71	0.81	30.5	22.1	8.4
B 34	85	HP	7.9	69	642	480.3	21.89	1.84	85.63	10.17	118.7	1.41	64.09	22.46	43.03	2.80	41.10	0.83	0.97	0.96	31.2	21.2	10.0
B 35	80	TW	8.0	67	522	399.0	13.01	1.09	67.68	11.14	166.2	0.79	23.56	29.58	13.36	2.91	35.32	0.75	1.06	1.75	29.9	17.4	12.5
B 36	90	HP	7.8	92	887	659.9	22.40	1.58	24.16	10.56	153.1	0.36	42.50	31.20	31.60	2.70	36.29	0.65	0.43	1.66	19.9	12.2	7.7

(HP-Hand Pump, OW- Open Well, TW- Tube Well)

The concentration of  $\text{SiO}_2$  varied from 14 to 49.31 mg/L in Pre-monsoon and 17.27 to 55.55 mg/L in Post-monsoon season (Table 7&8). This slight increment in concentration indicates possibility of aluminosilicate weathering in rainy season.

The concentration of Fe varied from 0.66 to 7.62 mg/L in Pre-monsoon and 0.43 to 6.84 mg/L in Post-monsoon season while value of Mn was significantly low in both season (Table 7 & 8). Distribution pattern of Fe for Pre-monsoon and Post-monsoon has been shown in fig. (8 c & d respectively). This higher concentration of Fe as well as low concentration of Mn gives an indication of reducing environment.

#### **Arsenic and its speciation**

Total arsenic concentrations in the groundwater varied from 19.1  $\mu\text{g/L}$  to 118  $\mu\text{g/L}$  in Pre-monsoon and 18.5  $\mu\text{g/L}$  to 113.5  $\mu\text{g/L}$  in Post-monsoon season. Distribution pattern of As for Pre-monsoon and Post-monsoon has been shown in fig. (8 a&b respectively). It was observed that places where arsenic concentration is high is found close to Ganges and its tributary Koshi (Ghugri) river. In the Pre-monsoon period, high concentration of arsenic was present in the shallow aquifers (HP) of Rannuchak (118  $\mu\text{g/L}$ ), Marwa (117  $\mu\text{g/L}$ ), Sabour (94  $\mu\text{g/L}$ ), Mahesitilakpur (88  $\mu\text{g/L}$ ) and Sultanganj (87  $\mu\text{g/L}$ ) areas of Bhagalpur district (Table 1). Similar observations were recorded in the Post-monsoon period with high arsenic in the shallow aquifers (HP) of Rannuchak (102  $\mu\text{g/L}$ ), Marwa (113  $\mu\text{g/L}$ ), Sabour (91  $\mu\text{g/L}$ ), Mahesitilakpur (83  $\mu\text{g/L}$ ) and Sultanganj (84  $\mu\text{g/L}$ ) (Table 2). Most of these regions are commonly found in the north and northwest part of the Bhagalpur district. In general 47 % of sampling location exceeds the 50  $\mu\text{g/L}$  concentration limit, but the concentration is still not too high which indicates nascent stage of arsenic enrichment.

However, in many areas of Lower Meghna Estuary, Bangladesh, more than 80% of wells (Ravenscroft et al. 2005) and 93 % of wells in Hajiganj upazila in southeast Bangladesh exceed the 50  $\mu\text{g/l}$  concentration limit (Jakarya et al. 1998). These observations suggest that the high concentrations of arsenic in the lower catchment of the Ganges river indicating the existence of multiple source areas and the likelihood of related mechanisms of mobilization across the entire Central Gangetic plain. The arsenic concentration less than 50  $\mu\text{g/L}$  in the central portion of the Bhagalpur district could be possible due to an accumulation of coarser sediment along a Holocene course of River Ganges (Ravenscroft et al. 2005). This indicates that how depositional environment and geological age are important factors in controlling arsenic mobilization (Bhattacharya et al. 2004).

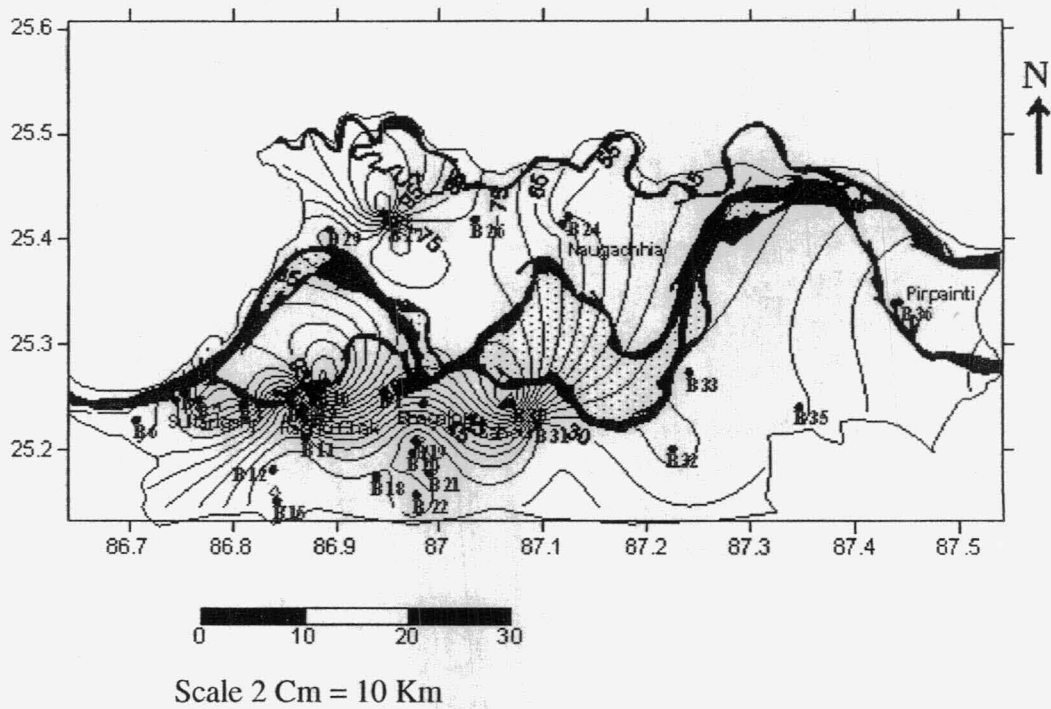


Fig :- 8(a) Contour map of Arsenic for Pre-monsoon in Bhagalpur

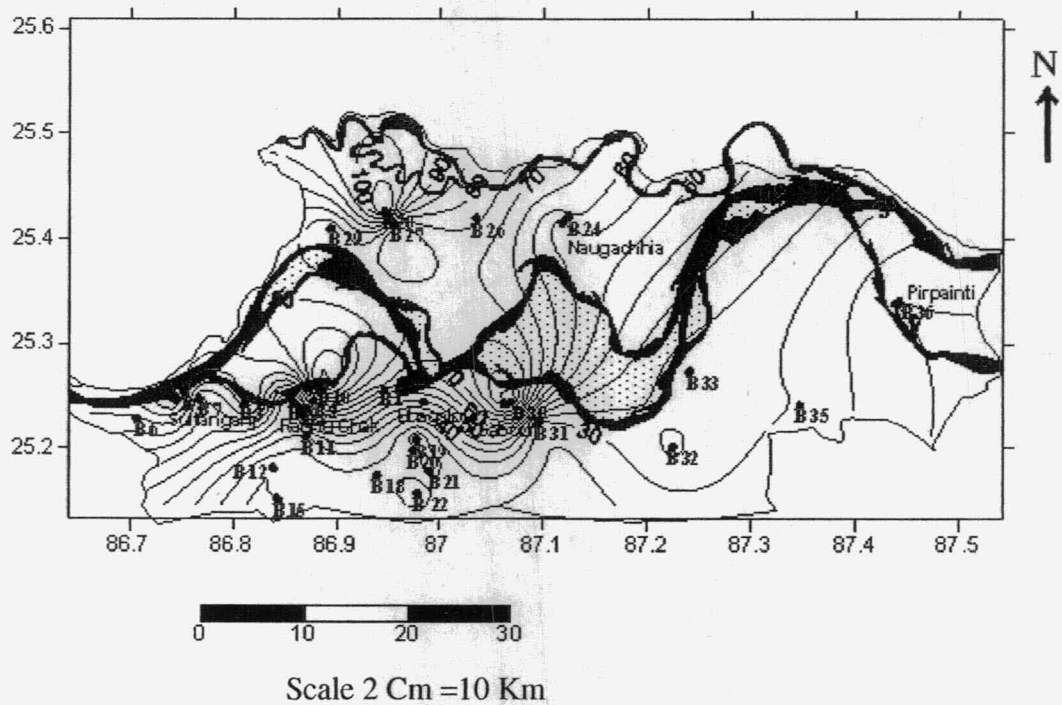


Fig :-8(b) Contour map of Arsenic for Post-monsoon in Bhagalpur

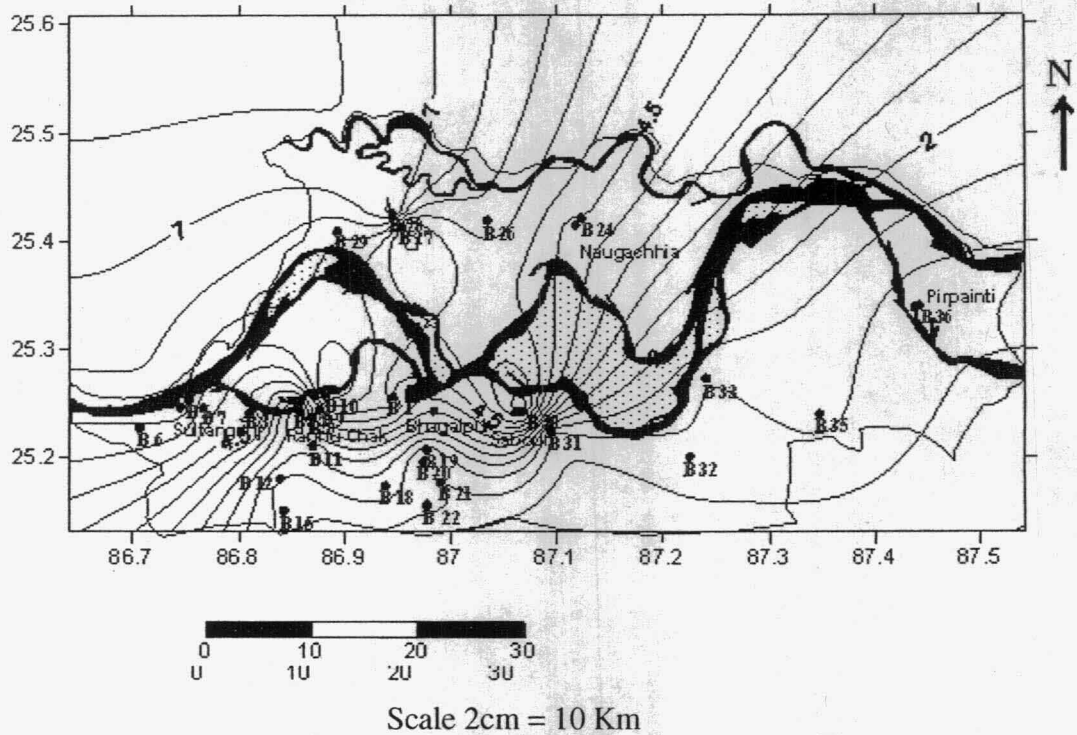


Fig.- 8(C) Contour map of Fe for pre monsoon in Bhagalpur

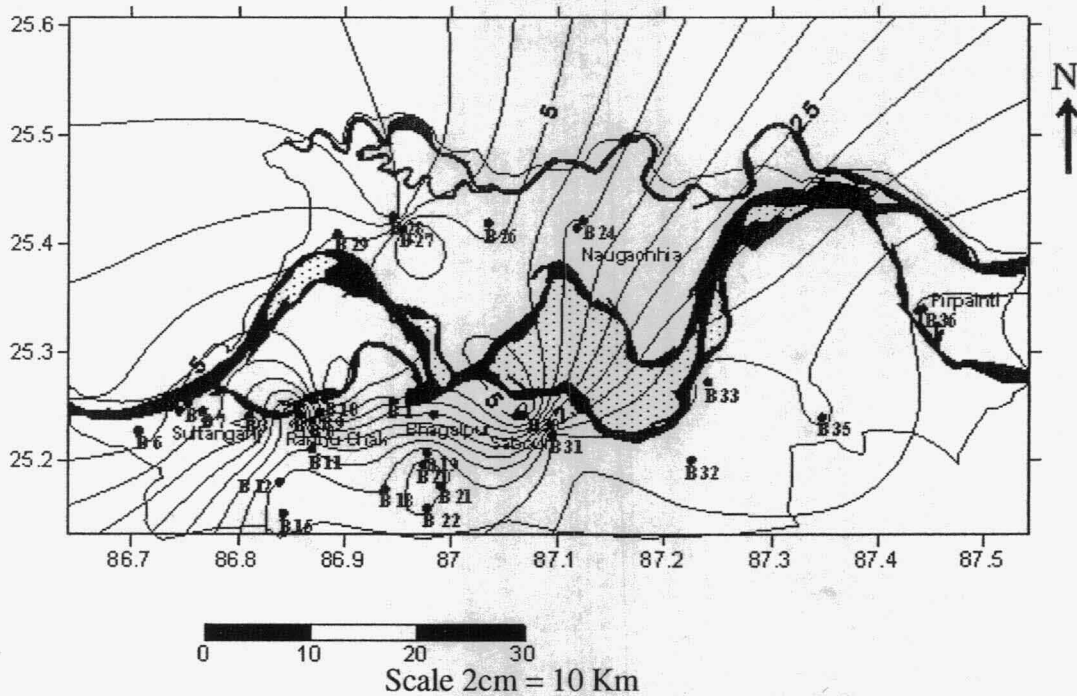


Fig.-8(d) Contour map of Fe for Post-monsoon in Bhagalpur

The depth distribution of arsenic shows strong negative correlation between occurrence of arsenic in groundwater and the depth of hand pumps or tube wells for both seasons shown in fig (10j & 11j). In general, the highest arsenic concentration

and also the expected spatial and temporal variability, occur 10-20 m below the ground surface, and decreases rapidly below about 40 meters. Similar observations are reported by (Karim et al. 1997) where a maximum arsenic concentration at 20 to 40 m depth, below which arsenic concentration decline. This requires strong redox conditions to drive arsenic mobility along the well depth (McArthur et al. 2004).

From different cross plots shown in fig. (10 d,e,f,g,h,i) and (11 d,e,f,g,h,i) it is clear that the high concentration of arsenic is associated with high concentration of Fe,  $\text{PO}_4^{3-}$  &  $\text{NH}_4^+$  as well as low concentration of Mn,  $\text{NO}_3^-$  and low value of Eh which favors development of anoxic condition during microbial oxidation of sedimentary organic matter along with some anthropogenic output i.e. it might have both the natural and artificial source. Low concentration of  $\text{NO}_3^-$  which reveals that the  $\text{NO}_3^-$  is thermodynamically favourable oxygen source for microbial degradation of dissolved organic materials in the shallow aquifers (Hand Pump) of Bhagalpur district. From all the above conditions we can conclude that "Oxyhydroxide reduction theory" is responsible for release of arsenic in aquifer of Bhagalpur.

To find the status of the aquifer here, we have also taken one core sample in the most effected region of Bhagalpur i.e. Rannu Chak upto 70 feet at the interval of 5 feet (shown in fig. 16) and we find that at depth of 20-30 feet, clay percentage is more and it may be responsible for reduced environment.

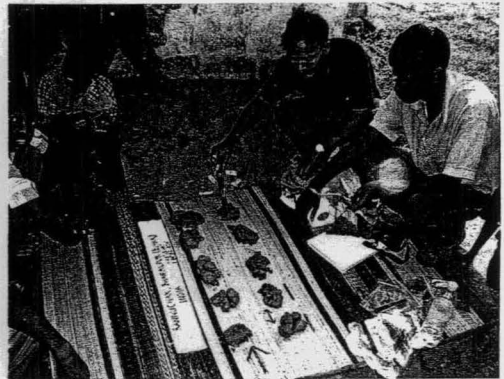
From arsenic speciation studies it has been observed that As (+3) Varies from varied from 10.9  $\mu\text{g/L}$  to 81.1  $\mu\text{g/L}$  in Pre-monsoon and 11.6  $\mu\text{g/L}$  to 75.9  $\mu\text{g/L}$  in Post-monsoon season (Table 7 & 8). The average value of  $\text{As}^{+3}$  is equals to 66-67 % of the average value of total As concentration, which is point of attention as it is prime source of lethality. Arsenic speciation shows that the proportion of arsenite ( $\text{As}^{+3}$ ) relative to arsenate ( $\text{As}^{+5}$ ) in aqueous samples are more. This indicate the shifting behaviour of river Ganga which may lead variation in hydrological budget which further trigger the reduction of oxidized As to reduced As, a more mobile and toxic species. It has been also found that  $\text{As}^{+3}$  showing same correlation as by the total As.

#### **Graphical representation of hydrochemical data**

In the present study, water quality data is analyzed through statistical distribution diagrams such as Piper diagram, Durov diagram and Schoeller digram to gain better insight into the hydrochemical processes operating in the groundwater

flow system that resulted in the observed spatial and temporal variation in the groundwater quality. Furthermore, the major cations and anions for the analyzed water were plotted on a Piper diagram. Hydrochemical diversity among the samples is revealed by the Piper diagram for Pre-monsoon and Post-monsoon seasons shown in fig. (10 a& 11 a respectively). It has been found that most of the water samples are of Ca-Cl (about 62%) and Ca-HCO<sub>3</sub> ( about 36%) type in Pre-monsoon period while in Post-monsoon period most of water samples are also Ca-HCO<sub>3</sub> and Ca-Cl type but in different ratios that are 59% and 40% respectively. This indicates presence of carbonaceous sandstones in the aquifers and weathering of carbonate minerals in Post-monsoon period while in the Pre-monsoon period result favours salt precipitation. In most hardness are of CaCO<sub>3</sub> type. Some of them are saturated with Calcite. From Durov diagram for Pre-monsoon and Post-monsoon shown in fig.(10b & 11b) it became clear that most of the water sample are of alkaline in nature and alkalinity decreases in Post-monsoon due to dilution effect. From Schoeller diagram for Pre-monsoon and Post-monsoon shown in fig. (10c & 11c) it became clear that Ca is dominant cation while Na & K are very less in value due to poor water- silicate rock interaction. The Post-monsoon samples indicate permanent hardness domain as well as ion exchange and dissolution processes occurring due to recharge of the groundwater in the monsoon period.





During Core drilling in Rannuchak, Bhagalpur, Bihar



With local drillers and my senior lab mate (left), Speciation of under groundwater during drilling

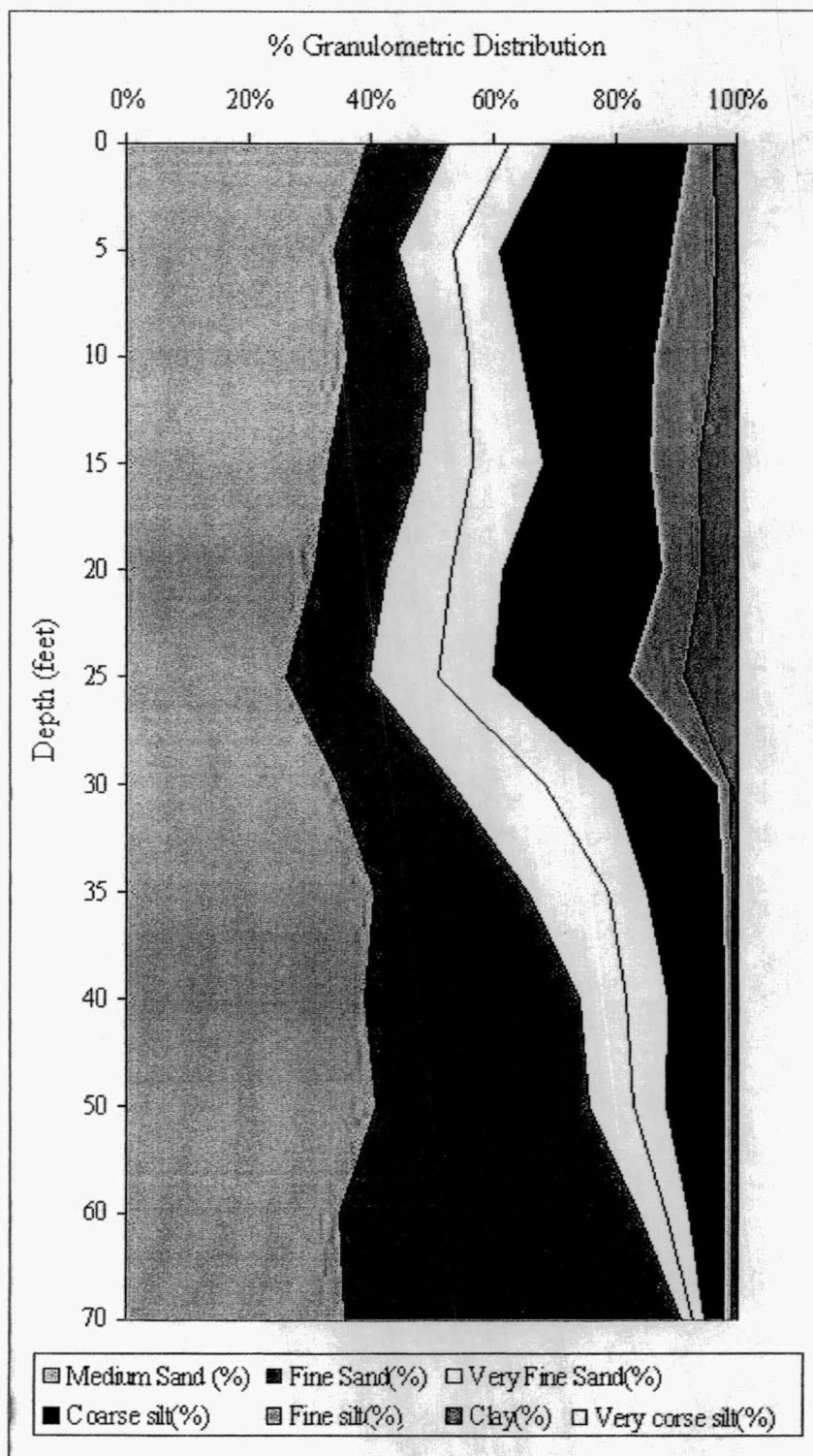


Figure 16- Vertical soil profile of Bhagalpur, Bihar

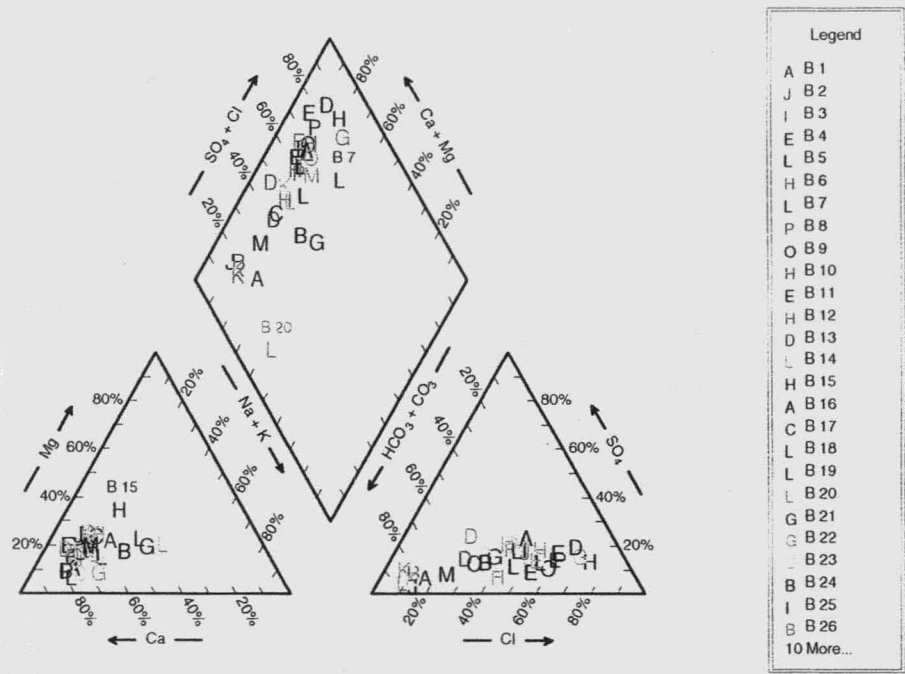


Fig. 10 (a) Piper diagram of Bhagalpur for Pre-Monsoon

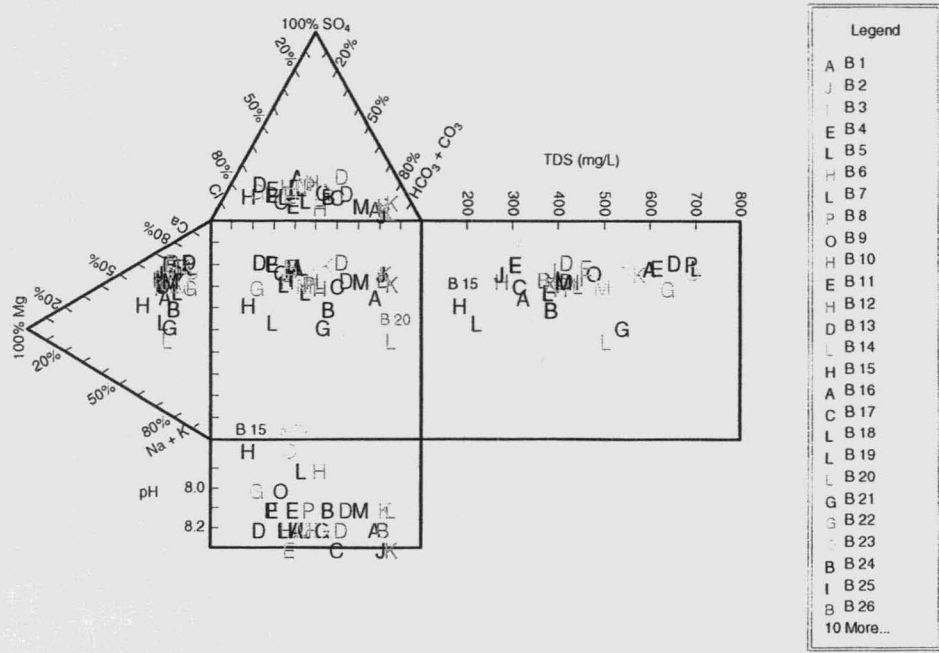


Fig. 10 (b) Durov diagram of Bhagalpur for Pre-Monsoon

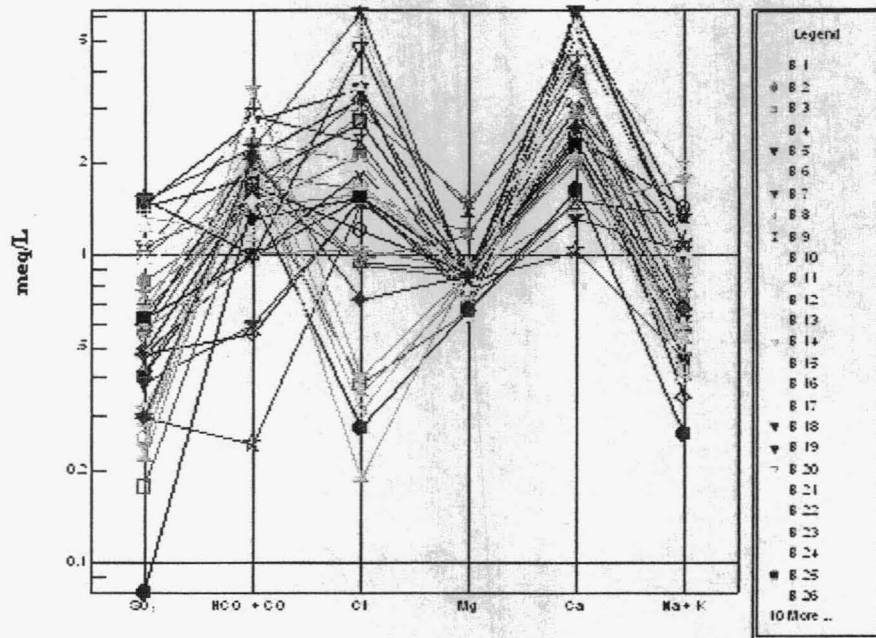


Fig. 10 (c) Schoeller diagram of Bhagalpur for Pre-Monsoon

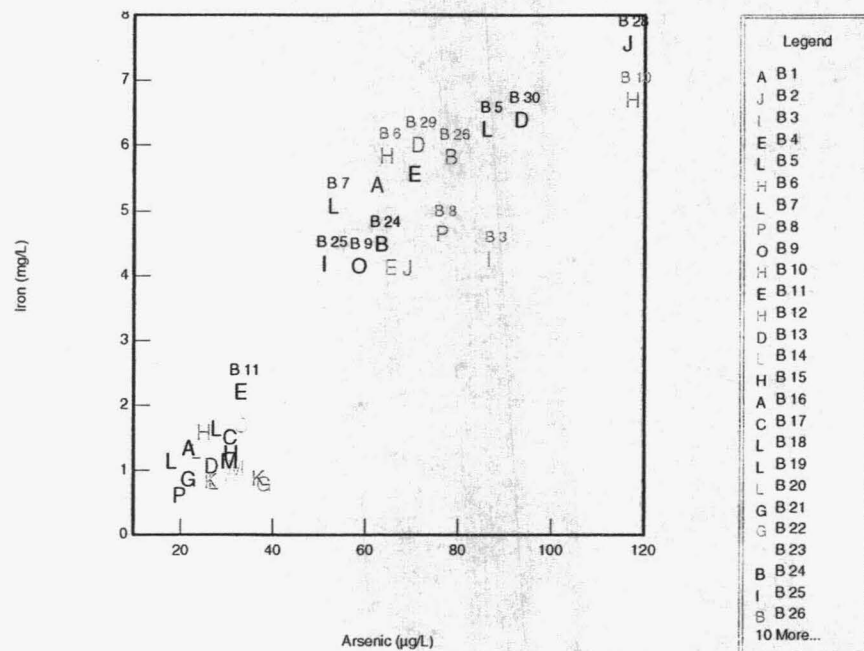


Fig. 10 (d) Scatter diagram of As vs. Fe of Bhagalpur for Pre-Monsoon

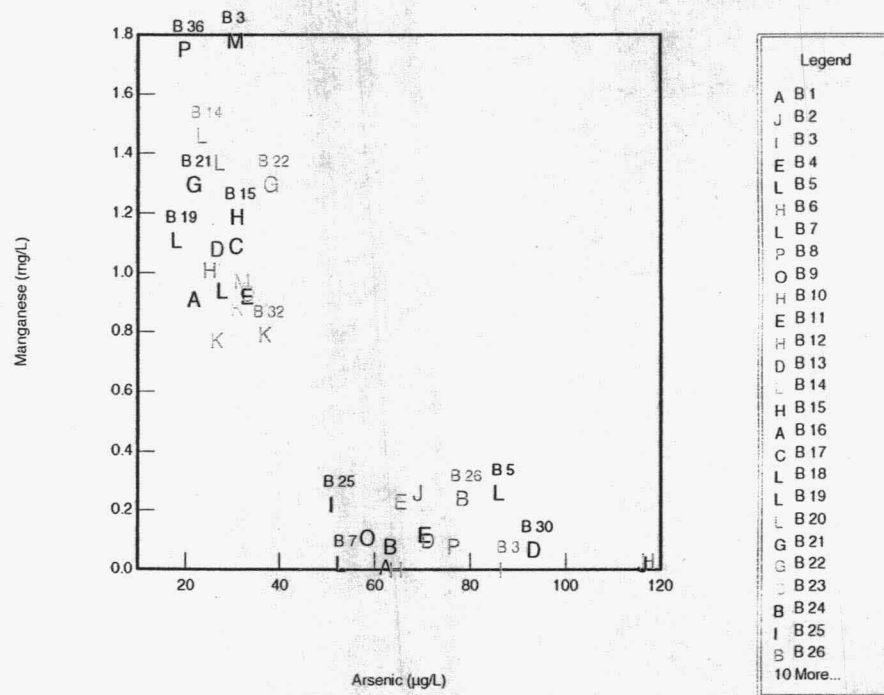


Fig. 10 (e) Scatter diagram of As vs. Mn of Bhagalpur for Pre-Monsoon

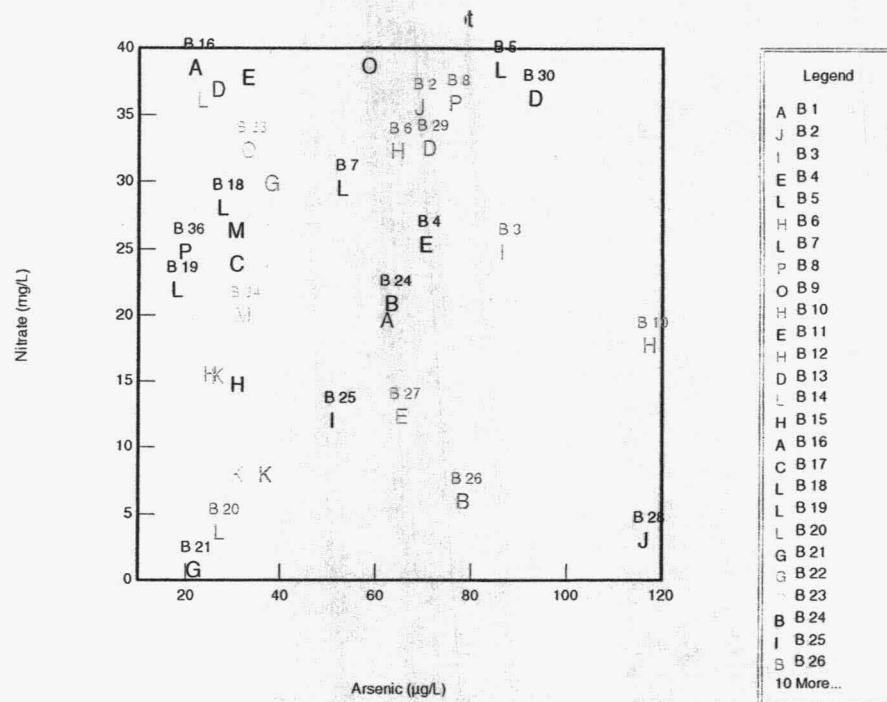


Fig. 10 (f) Scatter diagram of As vs. Nitrate of Bhagalpur for Pre-Monsoon

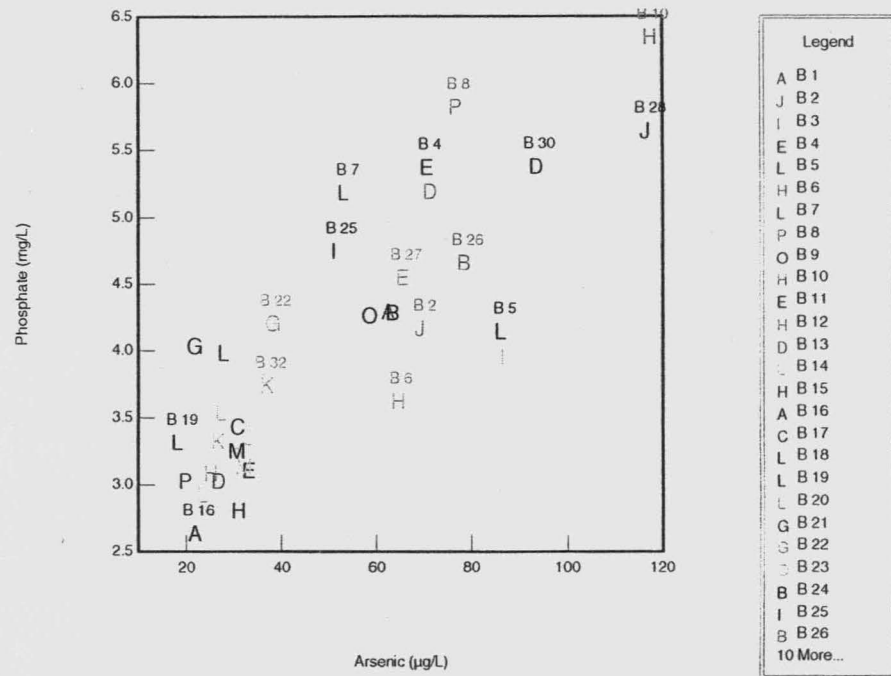


Fig. 10 (g) Scatter diagram of As vs. Phosphate of Bhagalpur for Pre-Monsoon

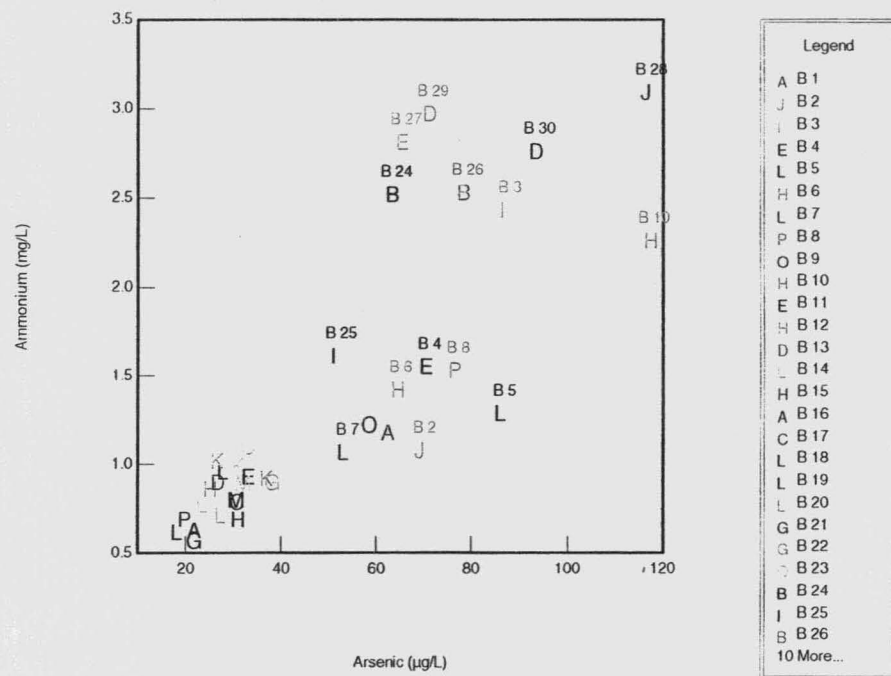


Fig. 10 (h) Scatter diagram of As vs. Ammonium of Bhagalpur for Pre-Monsoon

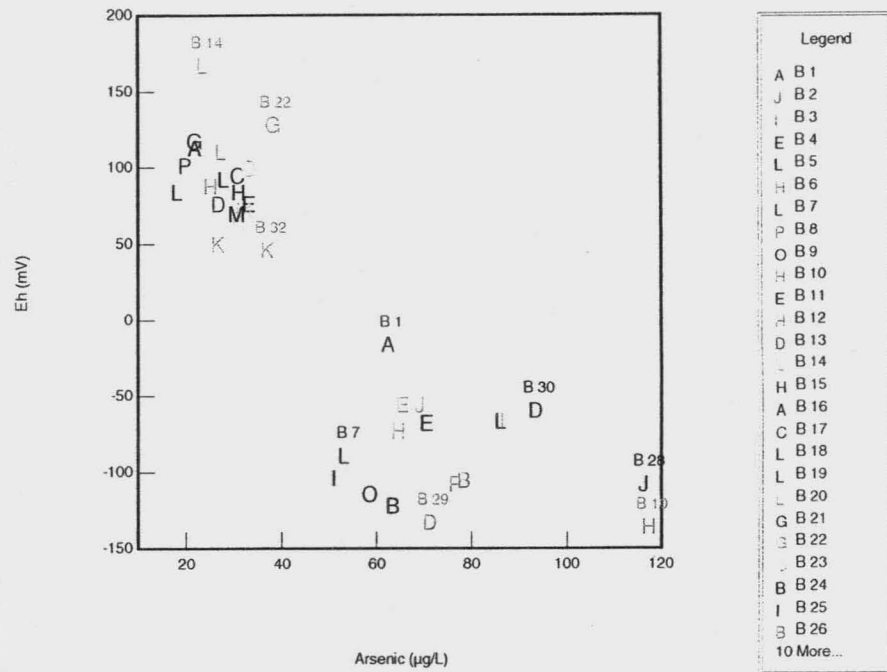


Fig. 10 (i) Scatter diagram of As vs. Eh of Bhagalpur for Pre-Monsoon

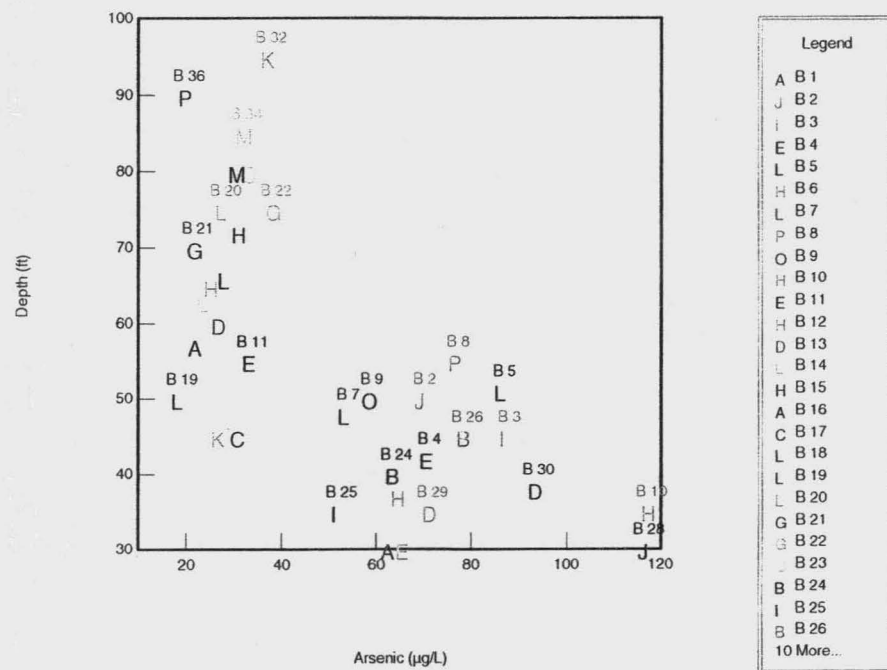


Fig. 10 (j) Scatter diagram of As vs. Depth of Bhagalpur for Pre-Monsoon

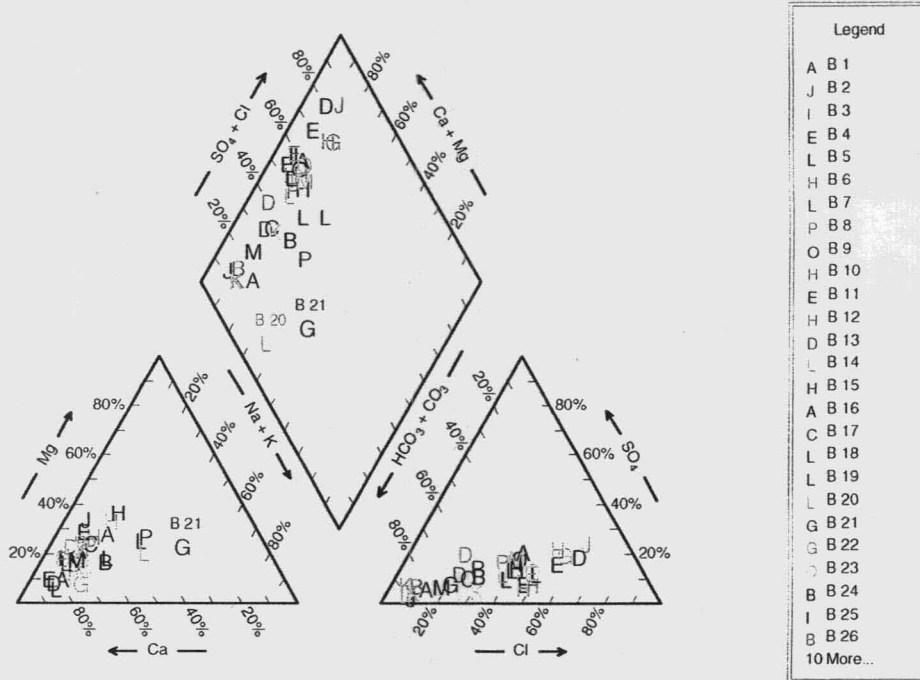


Fig. 11 (a) Piper diagram of Bhagalpur for Post-Monsoon

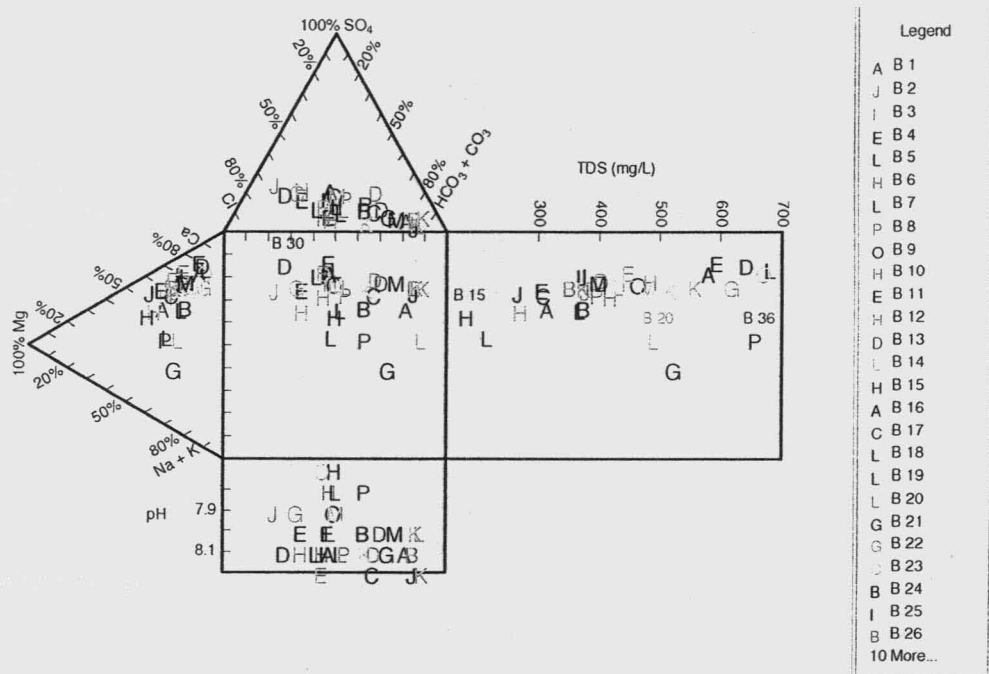


Fig. 11 (b) Durov diagram of Bhagalpur for Post-Monsoon



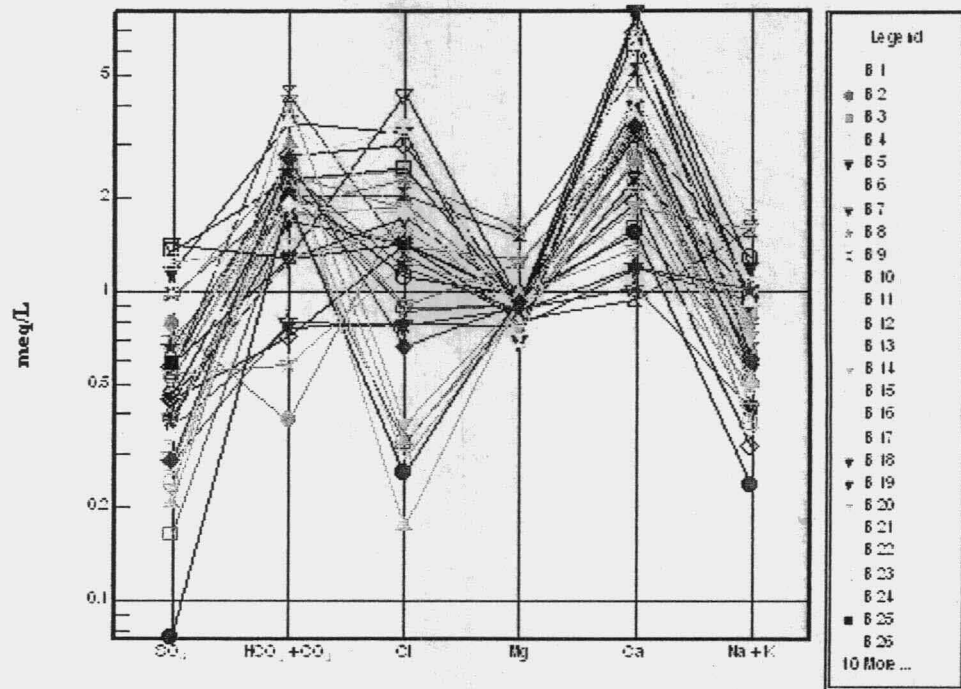


Fig. 11 (c) Schoeller diagram of Bhagalpur for Post-Monsoon

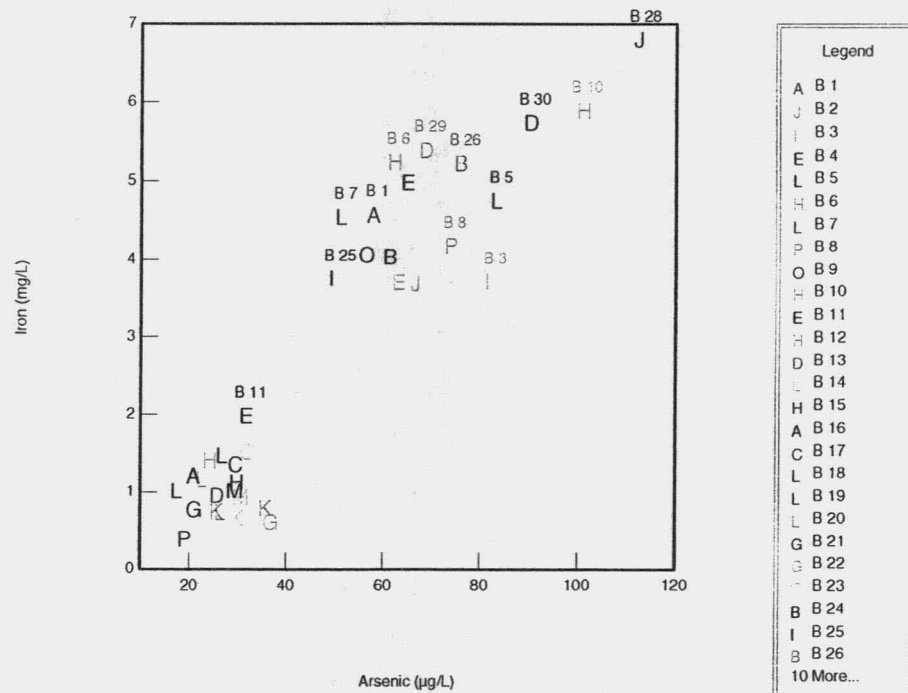


Fig. 11 (d) Scatter diagram of As vs. Fe of Bhagalpur for Post-Monsoon

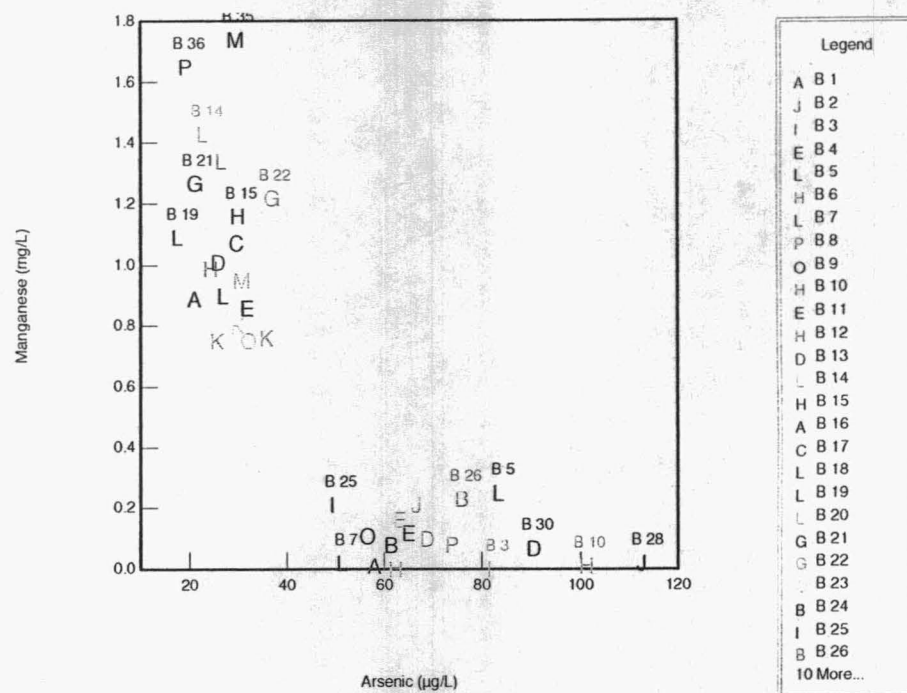


Fig. 11 (e) Scatter diagram of As vs. Mn of Bhagalpur for Post-Monsoon

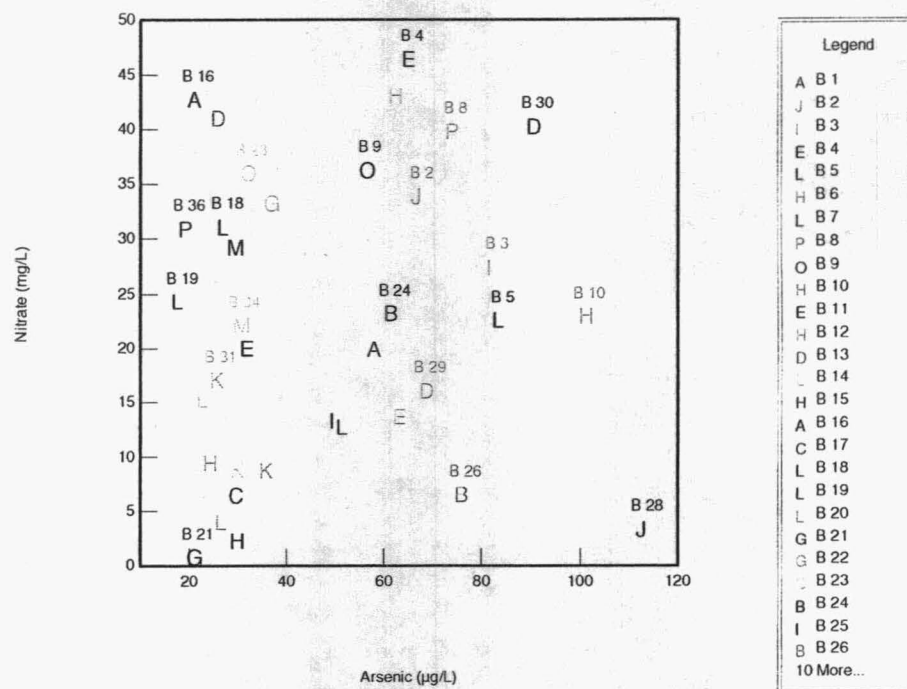


Fig. 11 (f) Scatter diagram of As vs. Nitrate of Bhagalpur for Post-Monsoon

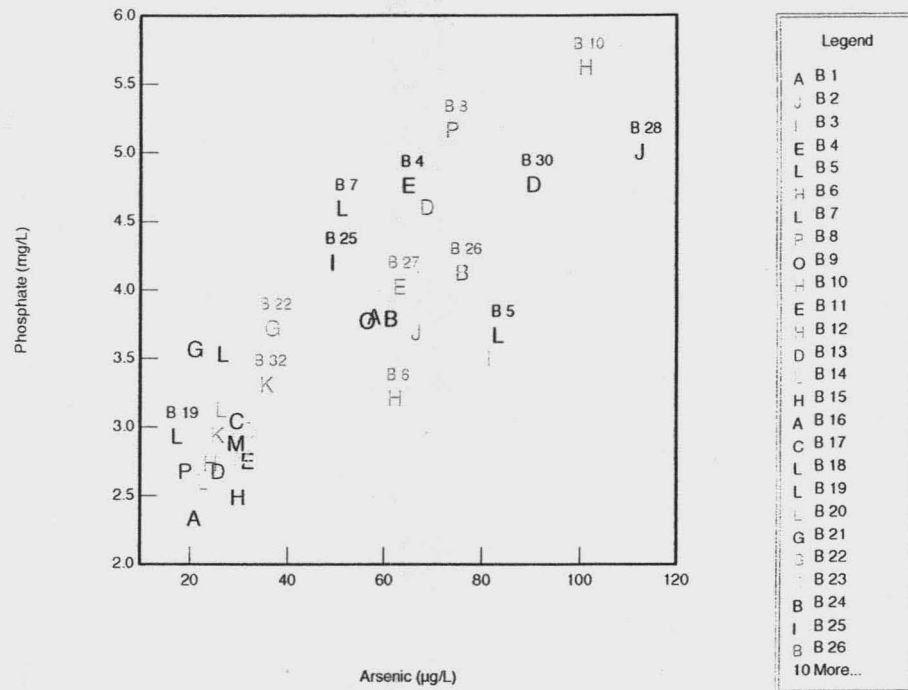


Fig. 11 (g) Scatter diagram of As vs. Phosphate of Bhagalpur for Post-Monsoon

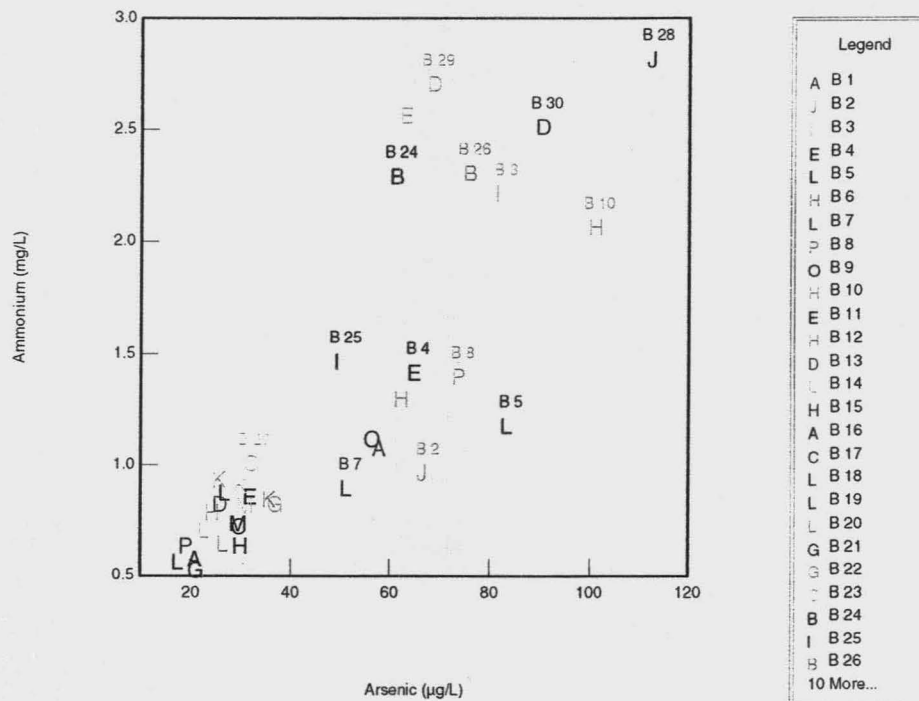


Fig. 11 (h) Scatter diagram of As vs. Ammonium of Bhagalpur for Post-Monsoon

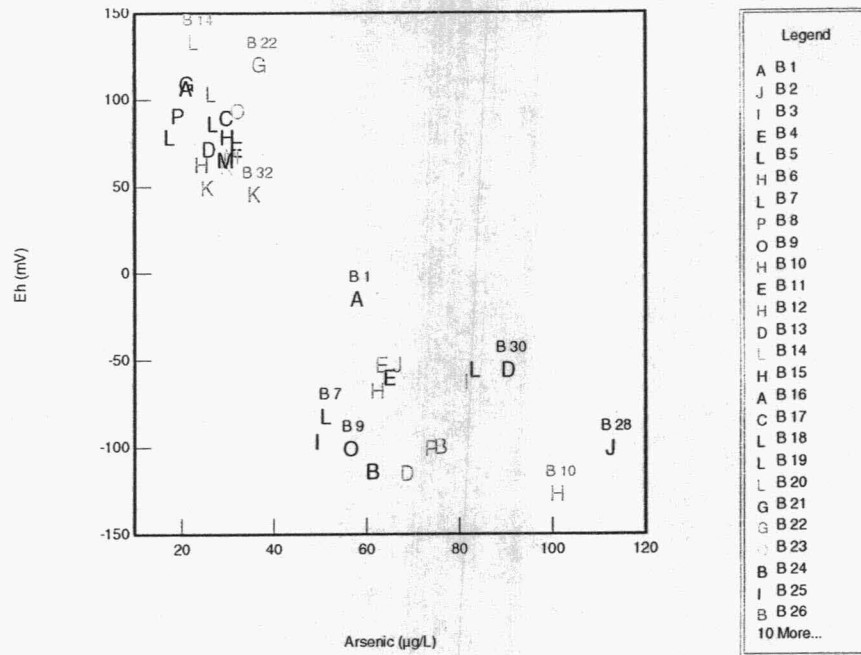


Fig. 11 (i) Scatter diagram of As vs. Eh of Bhagalpur for Post-Monsoon

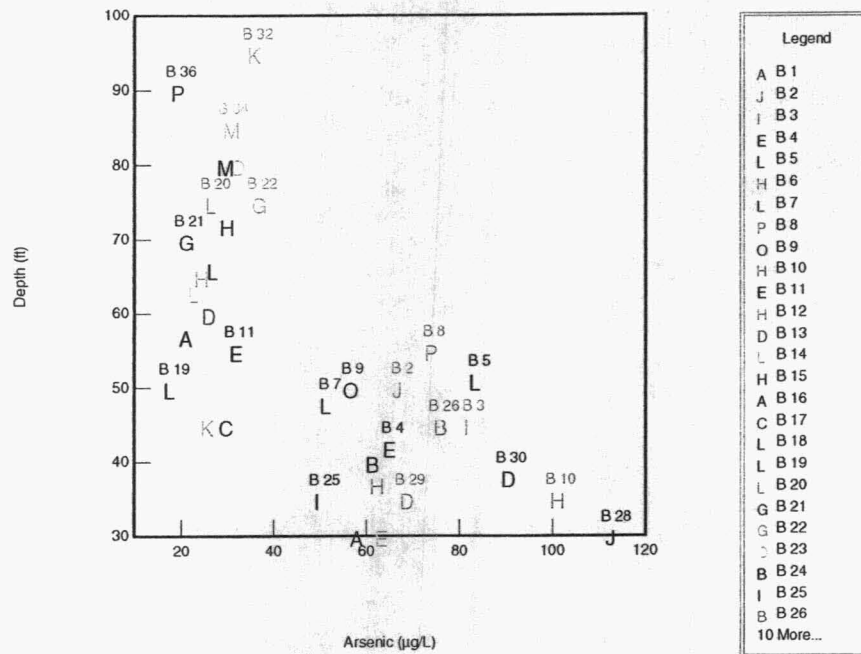


Fig. 11 (j) Scatter diagram of As vs. Depth of Bhagalpur for Post-Monsoon

### **Statistical analysis**

#### **Correlation analysis**

From table 3 and 4 we find correlation pattern for Pre-monsoon and Post-monsoon respectively which gives a clear picture of physio-chemical processes occurring their.

There is a good correlation of Na and K which indicates that source for both the element could be same. There are good correlation exists between TDS - Ca, TDS-Cl<sup>-</sup> & TDS - SO<sub>4</sub><sup>2-</sup> which indicates that carbonate weathering along with the anthropogenic sources mainly fertilizers control the geochemistry of Pre-monsoon ground water. There is also a good correlation found between Ca-Cl<sup>-</sup> & Ca-SO<sub>4</sub><sup>2-</sup> which may indicate that both gypsum (absorbed in clay) and limestone are acting as a source of calcium. During post monsoon season an additional correlation exist between TDS - HCO<sub>3</sub><sup>-</sup>, which indicates carbonate weathering. Other good correlation like between Cl<sup>-</sup> - SO<sub>4</sub><sup>2-</sup> & Cl<sup>-</sup> - NO<sub>3</sub><sup>-</sup> which may be result due to secondary leaching.

#### **Corelation related to Arsenic**

The corelation of arsenic and sulphate for Bhagalpur is very poor, which indicates arsenic is not released by oxidation of arsenic bearing pyrite or arseno-pyrite at rock / water / air interface. A good correlation (>0.5) between Sulphate with Ca and TDS for both seasons fig.(14) indicates that the acid produced from oxidation of pyrites is being neutralized by carbonate formed during weathering process, and arsenic released consequently possibly by sorbed in hydrated Fe-oxide also produced during weathering. High correlation between arsenic and phosphate shows the mobilization of arsenic may partially governed by anthropogenic agricultural activities.

Hydrated Fe-oxides produced on weathering have great capacity specifically for adsorbing or co-precipitating As are also produced during process. Reductive dissolution of hydrated iron oxide, which is likely to be mediated by Fe (III)-reducing bacteria, thus appears to have released Fe<sup>2+</sup> and sorbed As to groundwater. It has also been found that concentration of Fe and As are high in shallow aquifers. A positive co-relation has been noticed between As and Fe concentration high correlation justify the significance of this association.

There was an inverse correlation between As and Eh (Redox potential) i.e. value of Eh increases with the decrease in As concentration, which means concentration of As increases with the increment in reducing status of any environment. We found a good correlation of As with Fe,  $\text{NH}_4^+$  &  $\text{PO}_4^{3-}$  While inverse relation of As with depth, Mn, ORP &  $\text{NO}_3^-$  which indicates strong reducing character of this environment. This behavior of arsenic is governed by redox reactions at shallow depth which support the dissolution of arsenic from arseniferous iron oxyhydroxides. This assumption is being supported by high correlation of As – Fe.

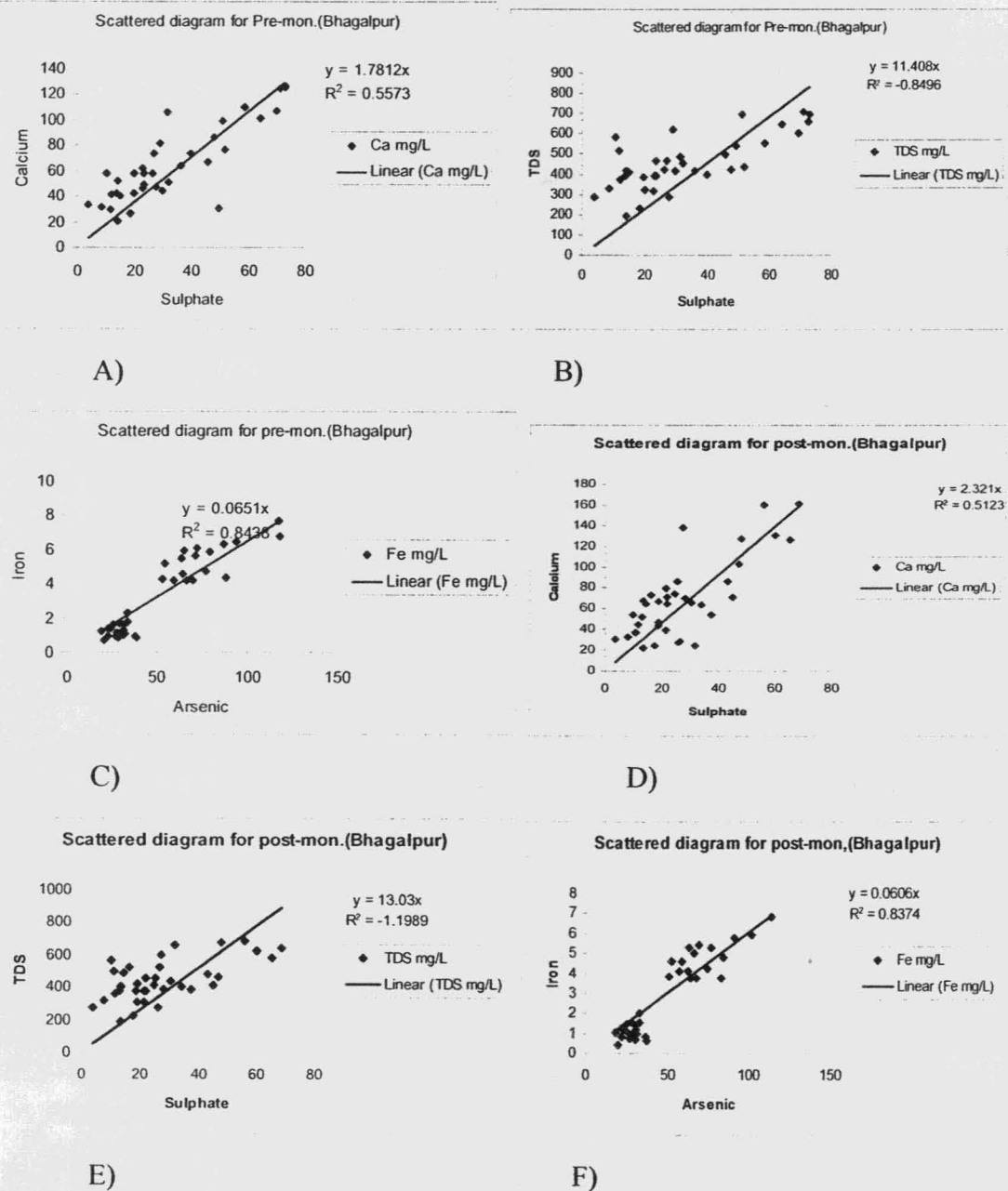


Fig. 14 (A to F) Different scattered diagrams showing relation between Calcium-Sulphate, TDS-Sulphate and Arsenic- Iron for both seasons in Bhagalpur.

### **Factor analysis**

Factor analysis of physico-chemical parameters of the Bhagalpur groundwater indicates trends for Pre-monsoon and post- monsoon periods. The total variability accounted for five factors is 80.38 % in Pre-monsoon and while it accounted for five factors is 81.77 % in post- monsoon. The number of significant factors within the data is established by considering only those with an Eigen value >1.0. The degree of association between each variable and each factor is given by its loading on that factor. The Pre-monsoon (Table 5) and post monsoon (Table 6) indicate the loading of each variable on each factor and the communality of each variable accounted for in the analysis.

In Pre-monsoon season, component 1 shows the strong geochemical associations between  $\text{PO}_4^{3-}$ , Fe, As and As(+3) and inverse association with Depth, ORP and Mn that are indicator of reducing environment which might be responsible for arsenic pollution in the water sample. Other parameter which also having strong effect on it is  $\text{SiO}_2$ , which indicates weathering of silicate minerals like feldspar. Component 2 represents the association with EC and TDS with some ions which that indicates percolation of salts and halite deposits. Component 3 is accounted for Na, K and  $\text{F}^-$  that indicate feldspar or biotite weathering. Component 4 explains the strong relationship of Ph with  $\text{HCO}_3^-$  that accounts for weathering activities.

In post monsoon season, all components show almost same association but with different intensity. Component 1 shows some extra association like with  $\text{NH}_4^+$  and Mn which indicate some biological inputs. For component 2 one new association with  $\text{NO}_3^-$  came which indicates fertilizer inputs. Component 4 shows a relation of EC and TDS with  $\text{HCO}_3^-$  which indicate weathering of carbonaceous material.

In Pre-monsoon and Post-monsoon the component factor analysis indicates the high association of As with Fe and water type is primarily govern by this iron-arsenic association. This mobilization of arsenic may be governed by redox reactions at shallow depth which support the dissolution of arsenic from arseniferous iron oxy-hydroxides.

Table 3 Bhagalpur Pre-monsoon corelation matrix (n=36)

	Depth	pH	ORP	EC	TDS	Na <sup>+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	HCO <sub>3</sub> <sup>--</sup>	F <sup>-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	PO <sub>4</sub> <sup>3-</sup>	SiO <sub>2</sub>	NH <sub>4</sub> <sup>+</sup>	Fe	Mn	As(tot)	As <sup>3+</sup>	As <sup>5+</sup>	
Depth	1																						
Ph	-0.21	1																					
ORP	0.71	-0.18	1																				
EC	0.23	-0.26	0.27	1																			
TDS	0.24	-0.23	0.28	0.98	1																		
Na <sup>+</sup>	0.36	-0.30	0.28	0.56	0.56	1																	
K <sup>+</sup>	0.37	-0.26	0.30	0.55	0.55	0.98	1																
Ca <sup>2+</sup>	0.13	-0.19	0.06	0.78	0.78	0.36	0.31	1															
Mg <sup>2+</sup>	-0.20	0.08	-0.48	-0.13	-0.11	-0.11	-0.13	0.15	1														
HCO <sub>3</sub> <sup>--</sup>	0.26	-0.01	0.27	0.54	0.53	0.38	0.41	0.17	-0.11	1													
F <sup>-</sup>	0.28	0.09	0.34	0.16	0.16	0.46	0.59	-0.22	-0.11	0.36	1												
Cl <sup>-</sup>	0.20	-0.33	0.16	0.70	0.70	0.50	0.48	0.78	0.04	-0.03	0.04	1											
NO <sub>3</sub> <sup>-</sup>	-0.09	-0.26	-0.04	0.13	0.12	0.07	0.01	0.42	0.17	-0.30	-0.28	0.37	1										
SO <sub>4</sub> <sup>2-</sup>	0.23	-0.14	0.22	0.73	0.73	0.49	0.49	0.85	0.06	0.15	0.09	0.81	0.27	1									
PO <sub>4</sub> <sup>3-</sup>	-0.59	0.28	-0.80	-0.12	-0.12	-0.11	-0.10	-0.04	0.43	-0.21	-0.08	0.02	-0.07	-0.04	1								
SiO <sub>2</sub>	0.19	-0.18	0.34	0.42	0.42	0.33	0.31	0.39	0.16	0.28	0.08	0.26	0.38	0.42	-0.28	1							
NH <sub>4</sub> <sup>+</sup>	-0.67	0.33	-0.78	-0.13	-0.12	-0.29	-0.31	0.02	0.26	-0.20	-0.31	-0.10	-0.12	-0.11	0.70	-0.29	1						
Fe	-0.78	0.21	-0.90	-0.27	-0.28	-0.30	-0.32	-0.09	0.30	-0.30	-0.26	-0.11	0.11	-0.21	0.81	-0.39	0.79	1					
Mn	0.78	-0.20	0.91	0.25	0.26	0.28	0.30	0.07	-0.33	0.33	0.34	0.17	-0.09	0.22	-0.74	0.31	-0.72	-0.89	1				
As(tot)	-0.67	0.28	-0.86	-0.22	-0.22	-0.22	-0.25	-0.03	0.37	-0.27	-0.24	-0.05	0.02	-0.16	0.83	-0.41	0.82	0.93	-0.89	1			
As <sup>3+</sup>	-0.66	0.20	-0.86	-0.17	-0.18	-0.16	-0.19	-0.03	0.37	-0.27	-0.20	-0.04	0.01	-0.13	0.85	-0.34	0.80	0.93	-0.81	0.95	1		
As <sup>5+</sup>	-0.44	0.34	-0.55	-0.23	-0.22	-0.27	-0.28	-0.02	0.24	-0.17	-0.22	-0.06	0.03	-0.15	0.50	-0.41	0.57	0.59	-0.57	0.75	0.50	1	



Table 4 Bhagalpur Post-monsoon correlation matrix (n=36)

	Depth	Ph	ORP	EC	TDS	Na <sup>+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	F <sup>-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	PO <sub>4</sub> <sup>3-</sup>	SiO <sub>2</sub>	NH <sub>4</sub> <sup>+</sup>	Fe	Mn	As(tot)	As <sup>3+</sup>	As <sup>5+</sup>	
Depth	1																						
Ph	-0.31	1																					
ORP	0.72	-0.24	1																				
EC	0.20	-0.33	0.26	1																			
TDS	0.22	-0.33	0.28	0.94	1																		
Na <sup>+</sup>	0.38	-0.29	0.32	0.54	0.54	1																	
K <sup>+</sup>	0.36	-0.28	0.29	0.52	0.52	0.91	1																
Ca <sup>2+</sup>	-0.06	-0.23	0.03	0.70	0.71	0.27	0.30	1															
Mg <sup>2+</sup>	-0.30	0.23	-0.55	-0.21	-0.20	-0.18	-0.16	-0.06	1														
HCO <sub>3</sub> <sup>-</sup>	0.27	-0.01	0.29	0.52	0.51	0.45	0.42	0.13	-0.13	1													
F <sup>-</sup>	0.34	0.02	0.38	0.15	0.16	0.60	0.60	-0.08	-0.14	0.34	1												
Cl <sup>-</sup>	-0.03	-0.31	0.05	0.66	0.67	0.37	0.39	0.85	-0.06	-0.08	-0.02	1											
NO <sub>3</sub> <sup>-</sup>	-0.06	-0.32	-0.11	0.49	0.50	0.12	0.13	0.65	0.13	-0.14	-0.12	0.61	1										
SO <sub>4</sub> <sup>2-</sup>	0.08	-0.26	0.14	0.61	0.62	0.41	0.42	0.77	-0.02	-0.05	0.04	0.78	0.54	1									
PO <sub>4</sub> <sup>3-</sup>	-0.59	0.39	-0.80	-0.12	-0.12	-0.10	-0.07	0.07	0.52	-0.17	-0.14	0.10	0.06	-0.03	1								
SiO <sub>2</sub>	0.50	-0.17	0.64	0.41	0.41	0.37	0.36	0.41	-0.27	0.19	0.25	0.41	0.25	0.46	-0.39	1							
NH <sub>4</sub> <sup>+</sup>	-0.67	0.37	-0.78	-0.11	-0.11	-0.31	-0.28	0.10	0.36	-0.16	-0.33	0.05	-0.02	-0.05	0.70	-0.55	1						
Fe	-0.79	0.26	-0.92	-0.24	-0.25	-0.34	-0.31	0.02	0.44	-0.33	-0.30	0.06	0.14	-0.10	0.82	-0.63	0.79	1					
Mn	0.76	-0.25	0.91	0.20	0.22	0.30	0.27	-0.09	-0.42	0.34	0.38	-0.09	-0.18	0.06	-0.74	0.56	-0.72	-0.89	1				
As(tot)	-0.67	0.32	-0.87	-0.21	-0.21	-0.26	-0.23	0.03	0.46	-0.29	-0.23	0.08	0.10	-0.11	0.82	-0.56	0.83	0.93	-0.83	1			
As <sup>3+</sup>	-0.65	0.27	-0.84	-0.17	-0.18	-0.28	-0.25	0.06	0.48	-0.24	-0.26	0.07	0.15	-0.15	0.84	-0.55	0.75	0.92	-0.83	0.92	1		
As <sup>5+</sup>	-0.47	0.30	-0.62	-0.20	-0.20	-0.14	-0.12	-0.02	0.28	-0.27	-0.11	0.07	0.00	-0.02	0.53	-0.40	0.68	0.64	-0.57	0.79	0.50	1	

Table 5- Factor analysis for Bhagalpur Pre-monsoon samples (n=36)

Variables	Comp.1	Comp.2	Comp.3	Comp.4	Comp.5	Factor	Eigen Value	Percentage of variance	Cumulative percentage
Depth	-0.738	0.133	0.233			1	7.407	33.668	33.668
Ph	0.244	-0.163	-0.208	0.649		2	4.658	21.175	54.843
ORP	-0.917	0.137	0.12		-0.128	3	2.697	12.257	67.1
EC	-0.147	0.909	0.24	0.106		4	1.62	7.365	74.465
TDS	-0.152	0.912	0.235	0.127		5	1.301	5.915	80.38
Na <sup>+</sup>	-0.145	0.434	0.81	-0.161					
K <sup>+</sup>	-0.159	0.399	0.865						
Ca <sup>2+</sup>		0.937	-0.107	-0.141	0.163				
Mg <sup>2+</sup>	0.388				0.756				
HCO <sub>3</sub> <sup>-</sup>	-0.266	0.231	0.461	0.586					
F <sup>-</sup>	-0.2		0.772	0.255					
Cl <sup>-</sup>		0.819	0.156	-0.369					
NO <sub>3</sub> <sup>-</sup>		0.264	-0.21	-0.683					
SO <sub>4</sub> <sup>2-</sup>		0.86	0.126	-0.115					
PO <sub>4</sub> <sup>3-</sup>	0.892								
NH <sub>4</sub> <sup>+</sup>	-0.376	0.362	0.101		0.692				
SiO <sub>2</sub>	0.843		-0.217	0.208					
Fe	0.948	-0.121	-0.116						
Mn	-0.891	0.127	0.151						
As(tot)	0.965								
As <sup>3+</sup>	0.945								
As <sup>5+</sup>	0.66		-0.23	0.136	-0.133				

Table 6 Factor analysis for Bhagalpur Post-monsoon samples (n=36)

Variables	Comp.1	Comp.2	Comp.3	Comp.4	Comp.5	Factor	Eigen Value	Percentage of variance	Cumulative percentage
Depth	-0.743		0.321			1	7.802	35.463	35.463
Ph	0.299	-0.269			0.85	2	4.608	20.948	56.41
ORP	-0.931		0.171			3	2.738	12.446	68.856
EC	-0.153	0.723	0.197	0.588	-0.132	4	1.703	7.743	76.599
TDS	-0.153	0.727	0.203	0.583	-0.112	5	1.139	5.176	81.775
Na <sup>+</sup>	-0.165	0.289	0.875	0.215	-0.141				
K <sup>+</sup>	-0.137	0.304	0.878	0.191	-0.134				
Ca <sup>2+</sup>		0.919		0.178					
Mg <sup>2+</sup>	0.518			-0.184	0.126				
HCO <sub>3</sub> <sup>-</sup>	-0.205		0.302	0.865					
F <sup>-</sup>	-0.21	-0.103	0.806		0.155				
Cl <sup>-</sup>		0.918	0.135						
NO <sub>3</sub> <sup>-</sup>	0.111	0.751			-0.284				
SO <sub>4</sub> <sup>2-</sup>		0.876	0.181	-0.101					
PO <sub>4</sub> <sup>3-</sup>	0.876				0.181				
NH <sub>4</sub> <sup>+</sup>	-0.622	0.516	0.196		0.289				
SiO <sub>2</sub>	0.844		-0.212	0.12	0.178				
Fe	0.952		-0.168						
Mn	-0.891		0.207	0.101					
As(tot)	0.956								
As <sup>3+</sup>	0.915		-0.125						
As <sup>5+</sup>	0.708			-0.205	0.194				

## **Geochemistry of Groundwater of Ghazipur, Uttar Pradesh**

The analytical precision for the measurement of ions was determined by calculating the Normalized inorganic Charge Balance (Huh et al. 1998), which is defined as  $[(Tz^+ - Tz^-) / (Tz^+ + Tz^-)]$  and represents the fractional difference between the total cations and total anions (Edmond et al. 1995). As exemplified by (Huh et al. 1998) the measured major ions ( $Na^+$ ,  $K^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Cl^-$ ,  $SO_4^{2-}$ ,  $HCO_3^-$ ,  $NO_3^-$ ) are generally enough to give a charge balance. Most of the ground water samples of Ghazipur showed a charge imbalance mainly in favor of positive charge. The observed average charge balance between cations ( $Tz^+$ ) and anions ( $Tz^-$ ) support the quality of the data points, which is  $\pm 11.69\%$  in Pre-monsoon where as it is  $9.76\%$  in Post-monsoon period. This imbalance of  $\Delta Tz$  could be related to the fact that no analysis was made on organic matter, which is mainly produced by biological activities during the Pre-monsoon and Post-monsoon (Berner-Kay and Berner 1987; Edmond et al. 1995; Huh et al. 1998).

The ground water was alkaline as evident from the average value of pH, which was 8.08 and 7.79 in the Pre-monsoon and Post-monsoon respectively (Table 15 & 16). The precipitation induced dissolution processes might have affected the pH of ground water. The high pH in the Pre-monsoon suggests that the soils/sediments are very reactive, which eventually enhances dissolution (Subramanian and Saxena 1983).

The value of EC varied from 318 to 1020  $\mu S/cm$  in the Pre-monsoon and 278.61 to 879  $\mu S/cm$  in the Post-monsoon (Table 15 & 16). The higher value of EC in the Pre-monsoon suggests the high concentration of dissolved solids and/or high ionic strength of groundwater, local variation in soil type, multiple aquifer system and agricultural activities in the area.

Bicarbonate represents the major source of alkalinity. In Ghazipur, bicarbonate concentration ranged between 48.8 to 262.3 mg/L in the Pre-monsoon and 60.79 to 326.77 mg/L in the Post-monsoon (Table 15 & 16). High  $HCO_3^-$  concentration in the Post-monsoon period indicates the weathering of carbonaceous sandstones in the aquifer, while low concentration in the Pre-monsoon period may be due to the precipitation of  $HCO_3^-$  along with other cations.

Variables	Unit	Minimum	Maximum	Average
Depth	(ft)	35.00	130.00	76.93
Ph		7.80	8.30	8.08
ORP	mv	-110.00	129.00	26.83
EC	$\mu\text{s}/\text{cm}$	318.00	1020.00	515.93
TDS	mg/L	233.66	749.46	379.70
Na <sup>+</sup>	mg/L	4.37	29.70	12.93
K <sup>+</sup>	mg/L	1.07	5.22	2.11
Ca <sup>2+</sup>	mg/L	42.50	121.81	87.78
Mg <sup>2+</sup>	mg/L	8.98	11.13	10.43
HCO <sub>3</sub> <sup>-</sup>	mg/L	48.80	262.30	120.98
F	mg/L	0.21	1.95	0.64
Cl	mg/L	2.57	288.49	49.46
NO <sub>3</sub> <sup>-</sup>	mg/L	1.28	76.77	21.31
SO <sub>4</sub> <sup>2-</sup>	mg/L	4.15	156.13	32.63
PO <sub>4</sub> <sup>3-</sup>	mg/L	2.19	5.25	3.46
NH <sub>4</sub> <sup>+</sup>	mg/L	11.96	42.16	25.86
SiO <sub>2</sub>	mg/L	0.54	2.84	1.35
Fe	mg/L	0.82	2.81	1.32
Mn	mg/L	1.02	1.47	1.14
As(tot)	$\mu\text{g}/\text{L}$	21.70	194.30	61.97
As <sup>3+</sup>	$\mu\text{g}/\text{L}$	15.50	138.78	41.86
As <sup>5+</sup>	$\mu\text{g}/\text{L}$	6.20	72.00	20.10

Table 15 – Statistical analysis of Pre-monsoon data of Ghazipur (n=30)

Variables	Unit	Minimum	Maximum	Average
Depth	(ft)	35.00	130.00	76.93
Ph		7.40	8.10	7.79
ORP	mv	-76.00	120.00	32.03
EC	$\mu\text{s}/\text{cm}$	278.61	879.00	452.61
TDS	mg/L	197.81	634.48	321.44
Na <sup>+</sup>	mg/L	3.83	25.98	11.31
K <sup>+</sup>	mg/L	0.87	4.24	1.71
Ca <sup>2+</sup>	mg/L	38.78	111.15	80.09
Mg <sup>2+</sup>	mg/L	8.66	10.74	10.06
HCO <sub>3</sub> <sup>-</sup>	mg/L	60.79	326.77	150.72
F	mg/L	0.23	1.90	0.63
Cl	mg/L	2.20	246.57	42.28
NO <sub>3</sub> <sup>-</sup>	mg/L	1.04	41.14	15.14
SO <sub>4</sub> <sup>2-</sup>	mg/L	3.83	139.87	27.10
PO <sub>4</sub> <sup>3-</sup>	mg/L	2.47	5.91	3.86
NH <sub>4</sub> <sup>+</sup>	mg/L	13.52	47.65	28.80
SiO <sub>2</sub>	mg/L	0.53	2.77	1.27
Fe	mg/L	0.90	3.10	1.46
Mn	mg/L	0.65	1.30	1.00
As(tot)	$\mu\text{g}/\text{L}$	18.76	155.40	52.31
As <sup>3+</sup>	$\mu\text{g}/\text{L}$	13.35	119.52	36.06
As <sup>5+</sup>	$\mu\text{g}/\text{L}$	5.42	52.51	16.26

Table 16 – Statistical analysis of Post-monsoon data of Ghazipur (n=30)

The  $\text{SO}_4^{2-}$  concentration varied from 4.15 to 156.13 mg/L in Pre-monsoon and 3.83 to 139.87 mg/L in Post-monsoon (Table 15&16). This indicates that the sulfate enrichment is high in Pre-monsoon due to breakdown of organic material and agriculture runoff carrying unutilized  $\text{SO}_4^{2-}$  (Anderson et al. 1979). The groundwater may have secondary salinity, as indicated by high  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  concentrations.

In this study,  $\text{Cl}^-$  varied from 2.57 to 288.49 mg/L in the Pre-monsoon and 2.20 to 246.57 mg/L in the Post-monsoon (Table 15 & 16). The higher and lower concentration of  $\text{Cl}^-$  in the Pre-monsoon and post monsoon respectively may be due to the input from sewage effluents in the village areas and dilution by rain water in post monsoon season (Todd 1959). The physical foresees such as mixing with another aquifer of different  $\text{Cl}^-$  concentrations and change in evaporation rate during recharge either spatially or temporally may cause the seasonal changes in the chloride content of groundwater. The high concentrations of  $\text{Cl}^-$  suggests mixing of fresh groundwater with soil water rich in dissolved ions derived from the salt precipitate coating on soils after Pre-monsoon season i.e. may be leaching during monsoon season. The presence of salts in unsaturated top zone of groundwater suggests that the flushing rate of the aquifers may be slow due to high  $\text{Cl}^-$  concentration trapped in clayey lenses, which may be gradually diffusing into the aquifer.

In Ghazipur,  $\text{NO}_3^-$  varied significantly from 1.28 to 76.77 mg/L in Pre-monsoon and 1.04 to 41.14 mg/L in Post-monsoon (Table 15 & 16), which indicates the contact of groundwater to agricultural activities and microbial mineralization. Distribution pattern of it for Pre-monsoon and Post-monsoon has been shown in fig. (9 e & f) respectively.

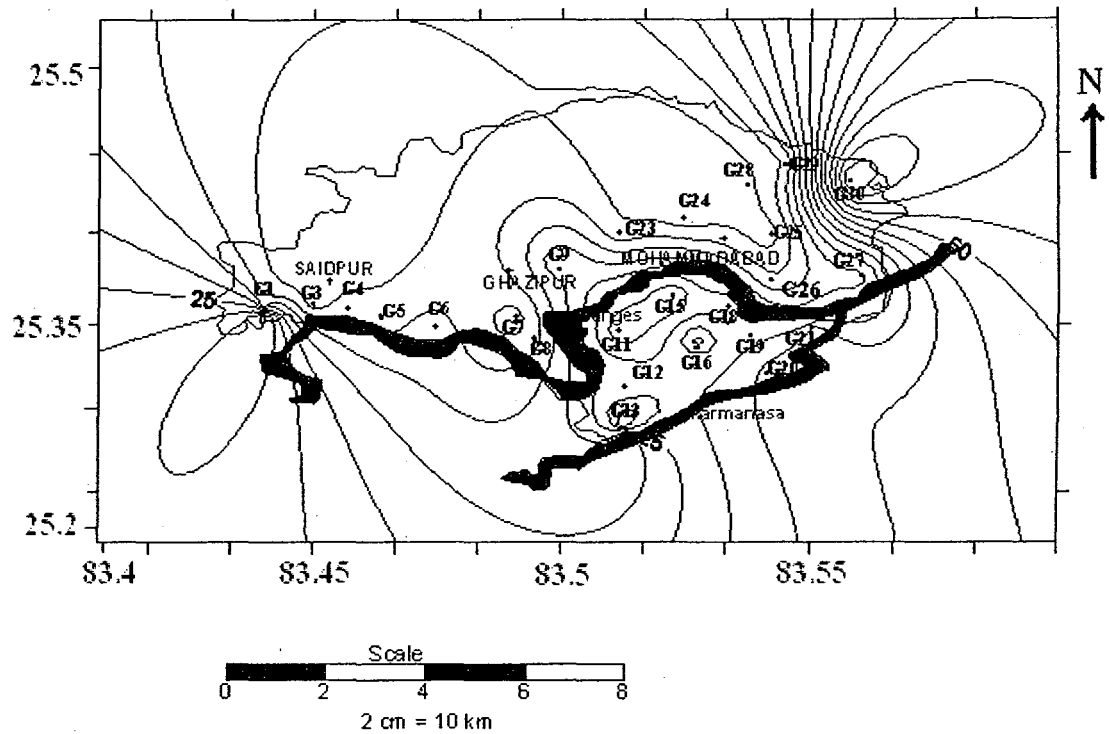


Fig 9(e)- Contour Map of Nitrate for Pre-monsoon in Ghazipur

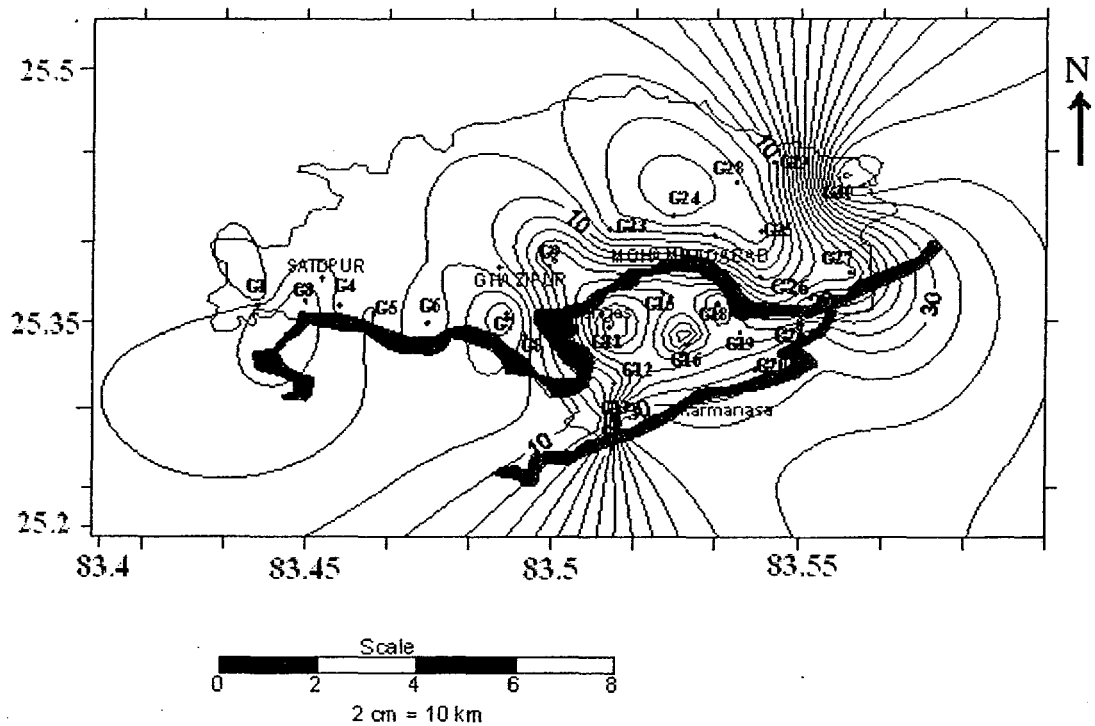


Fig 9(f)- Contour Map of Nitrate for Post-monsoon in Ghazipur

The concentration of  $PO_4^{3-}$  varied from 2.19 to 5.25 mg/L in the Pre-monsoon while in the Post-monsoon it ranged between 2.47 to 5.91 mg/L (Table 15 & 16). The underlying cause of  $PO_4^{3-}$  in groundwater of Ghazipur district indicates input of

fertilizers in farmlands to enhance the paddy and wheat productivity in rainy season. It also act as secondary leachates.

In Ghazipur, F<sup>-</sup> present in very insignificant amount and the concentration varies from 0.21 to 1.95 mg/L in Pre-monsoon and 0.23 to 1.90 mg/L in Post-monsoon (Table 15 & 16), which is indicator of weathering of mica containing mineral like biotite.

Finally among anions bicarbonate is the dominant species while average value trend found was HCO<sub>3</sub><sup>-</sup> > Cl<sup>-</sup> > SO<sub>4</sub><sup>2-</sup> > NO<sub>3</sub><sup>-</sup> > PO<sub>4</sub><sup>3-</sup> > F<sup>-</sup> in both period.

The dominant cation was Ca<sup>2+</sup> followed by Na<sup>+</sup>, Mg<sup>2+</sup> K<sup>+</sup> and NH<sub>4</sub><sup>+</sup>. There is a slight variation in seasonal distribution of all the ions and are significant at certain locations. All major cations show a particular trend with high concentration in Pre-monsoon and low in Post-monsoon season due to dilution effect. The concentration of Ca<sup>2+</sup> varied from 42.5 to 121.8 mg/L in Pre-monsoon and 38.78 to 111.15 mg/L in Post-monsoon season (Table 15 & 16). In Pre-monsoon season high concentration of Ca<sup>2+</sup> may be due to weathering of carbonate and plagioclase feldspar minerals, which is abundant in flood plain regions (Bhattacharya et al., 1997). In the Pre-monsoon the Mg<sup>2+</sup> concentration varied from 8.98 to 11.13 mg/L while in the Post-monsoon it was ranged between 8.66 to 10.74 mg/L (Table 15 & 16).

In the Pre-monsoon the Na<sup>+</sup> concentration varied from 4.37 to 29.7 mg/L while in the Post-monsoon it was ranged between 3.83 to 25.98 mg/L (Table 15 & 16). This infers that the contribution of cations via alumino-silicate weathering is low in comparison to carbonate weathering. The average ratio trend of Ca<sup>2+</sup>+Mg<sup>2+</sup>/Na<sup>+</sup>+K<sup>+</sup> varied from 6.53 in the Pre-monsoon and 6.92 in the Post-monsoon indicates the dominance of carbonate rock weathering in the Ghazipur ground water.

The concentration of NH<sub>4</sub><sup>+</sup> varied from 0.54 to 2.84 mg/L in Pre-monsoon and 0.53 to 2.77 mg/L in Post-monsoon season (Table 15 & 16). This higher concentration indicates a possible contamination from surface-water sources.

The concentration of SiO<sub>2</sub> varied from 11.96 to 42.16 mg/L in Pre-monsoon and 13.52 to 47.65 mg/L in Post-monsoon season (Table 15 & 16). This slight increment in concentration indicates possibility of alumino-silicate weathering in rainy season.

The concentration of Fe varied from 0.82 to 2.81 mg/L in Pre-monsoon and 0.90 to 3.10 mg/L in Post-monsoon season while value of Mn was significantly low in



both season (Table 15&16). Distribution pattern of Fe for Pre-monsoon and Post-monsoon has been shown in fig. (9c&d respectively) This higher concentration of Fe as well as low concentration of Mn gives an indication of reducing environment.

#### **Arsenic and its speciation**

Total arsenic concentrations in the groundwater varied from 21.7  $\mu\text{g/L}$  to 194.3  $\mu\text{g/L}$  in Pre-monsoon and 18.76  $\mu\text{g/L}$  to 155.4  $\mu\text{g/L}$  in Post-monsoon season as shown in (Table 9&10). Distribution pattern of As for Pre-monsoon and Post-monsoon has been shown in fig.(9 a&b respectively). It was observed that places where arsenic concentration is high is found close to Ganges and its tributary Karmnasa river. In the Pre-monsoon period, high concentration of arsenic was present in the shallow aquifers (HP) of Devbaranpur (194.3  $\mu\text{g/L}$ ), Jamania (177  $\mu\text{g/L}$ ), Gahmar (127  $\mu\text{g/L}$ ), Dildar Nagar (122  $\mu\text{g/L}$ ), Fulli (78.3  $\mu\text{g/L}$ ), Karanda (76.7  $\mu\text{g/L}$ ), Mohammadabad (73.2  $\mu\text{g/L}$ ) and Reotipur (70.2  $\mu\text{g/L}$ ) areas of Ghazipur district (Table 9). Similar observations were recorded in the Post-monsoon period with high arsenic in the shallow aquifers (HP) of Devbaranpur (155.4  $\mu\text{g/L}$ ), Jamania (142.6  $\mu\text{g/L}$ ), Gahmar (110.1  $\mu\text{g/L}$ ), Dildar Nagar (87.6  $\mu\text{g/L}$ ), Fulli (67.6  $\mu\text{g/L}$ ), Karanda (66.3  $\mu\text{g/L}$ ), Mohammadabad (63.3  $\mu\text{g/L}$ ) and Reotipur (61.0  $\mu\text{g/L}$ ) (Table 10). Most of these regions are commonly found in the south and southeast part of the Ghazipur district. In general 30 % of sampling location exceeds the 50  $\mu\text{g/L}$  concentration limit, but the concentration is still not too high which indicates nascent stage of arsenic enrichment.

However, in many areas of Lower Meghna Estuary, Bangladesh, more than 80% of wells (Ravenscroft et al. 2005) and 93 % of wells in Hajiganj upazila in southeast Bangladesh exceed the 50  $\mu\text{g/l}$  concentration limit (Jakarya et al. 1998). These observations suggest that the high concentrations of arsenic in the lower catchment of the river Ganga indicating the existence of multiple source areas and the likelihood of related mechanisms of mobilization across the entire Central Gangetic plain. The arsenic concentration less than 50  $\mu\text{g/L}$  in the central portion of the Ghazipur district could be possible due to an accumulation of coarser sediment along a Holocene course of River Ganges (Ravenscroft et al. 2005). This indicates that how depositional environment and geological age are important factors in controlling arsenic mobilization.

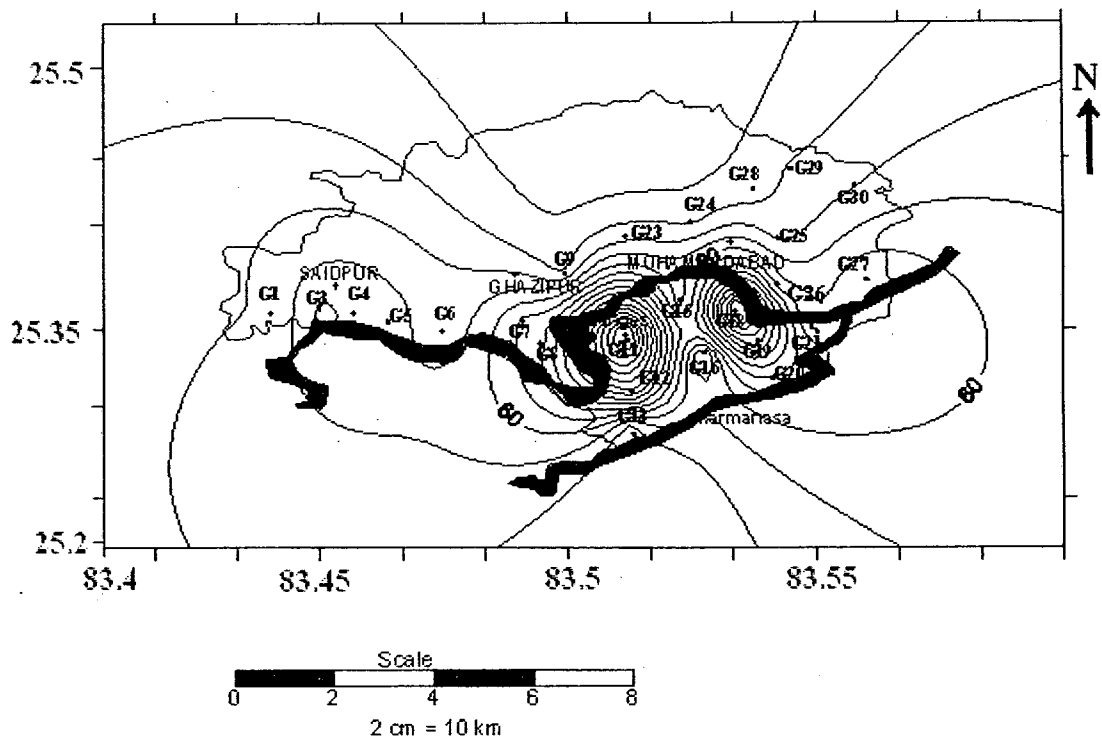


Fig 9(a)- Contour Map of Arsenic for Pre-monsoon in Ghazipur

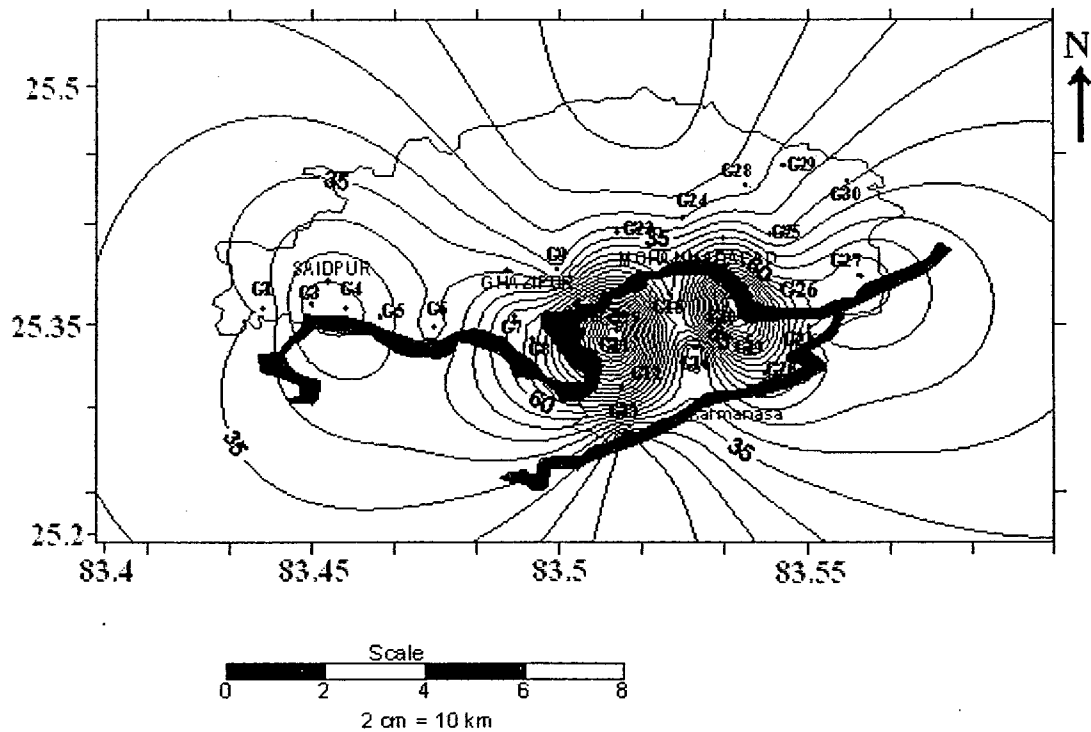


Fig 9(b)- Contour Map of Arsenic for Post-monsoon in Ghazipur

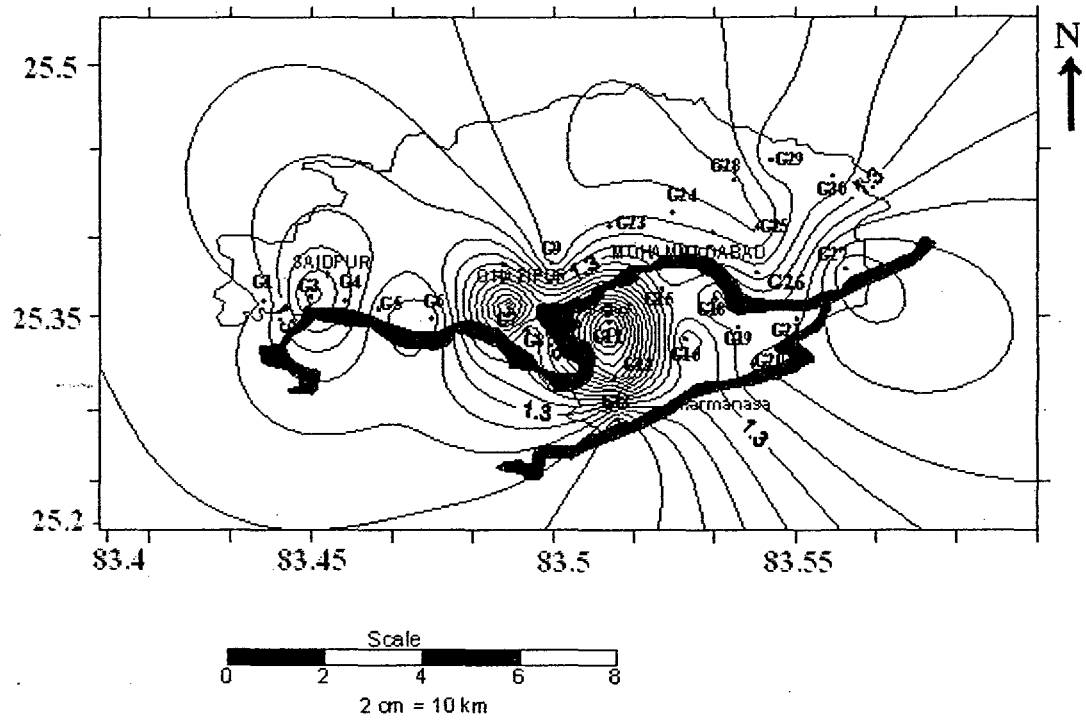


Fig 9(c)- Contour Map of Fe for Pre-monsoon in Ghazipur.

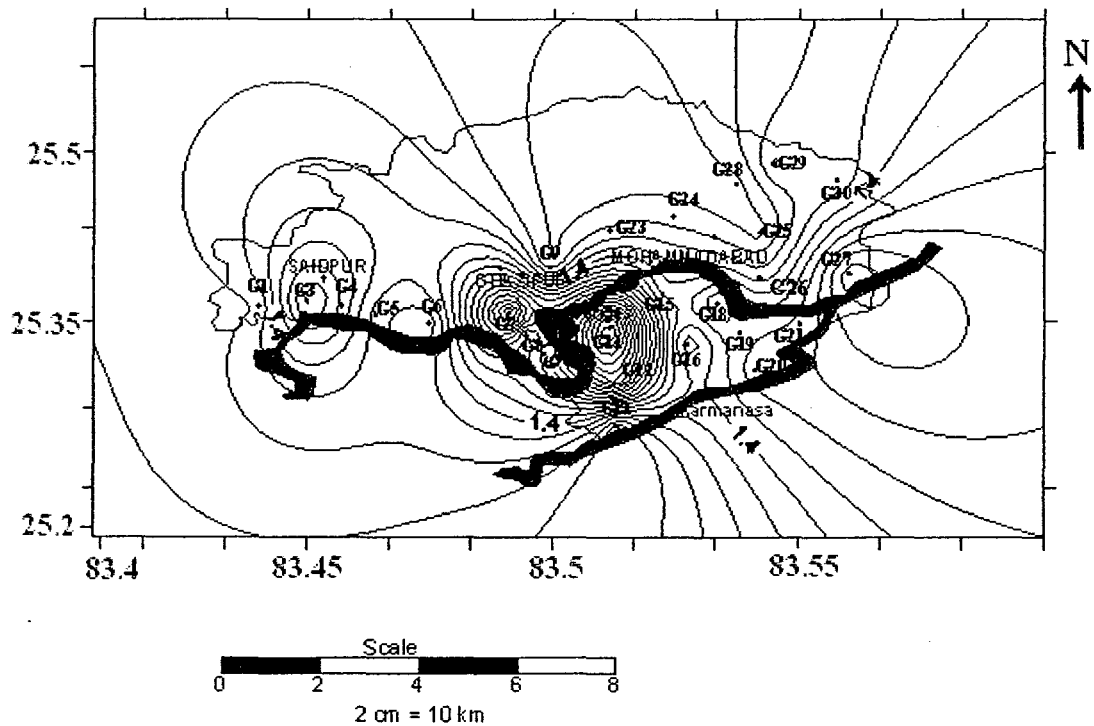


Fig 9(d)- Contour Map of Fe for Post-monsoon in Ghazipur.

Table 9 Hydro geochemistry of samples in Pre-monsoon of Ghazipur (n=30)

ID	Type	Depth (ft)	Ph	ORP Mv	EC μs/cm	TDS mg/L	Na <sup>+</sup> mg/L	K <sup>+</sup> mg/L	Ca <sup>2+</sup> mg/L	Mg <sup>2+</sup> mg/L	HCO <sub>3</sub> <sup>-</sup> mg/L	F <sup>-</sup> mg/L	Cl <sup>-</sup> mg/L	NO <sub>3</sub> <sup>-</sup> mg/L	SO <sub>4</sub> <sup>2-</sup> mg/L	PO <sub>4</sub> <sup>3-</sup> mg/L	SiO <sub>2</sub> mg/L	NH <sub>4</sub> <sup>+</sup> mg/L	Fe mg/L	Mn mg/L	As(tot) μg/L	As <sup>3+</sup> μg/L	As <sup>5+</sup> μg/L
G1	HP	85	8.2	116	700	514.3	16.12	2.69	94.5	9.17	128.1	0.6	101	43.25	51.57	3.56	15.74	1.10	1.09	1.04	41.4	29.6	11.8
G2	TW	120	8.3	104	650	477.6	14.28	2.38	98.77	9.57	115.9	1.07	89.1	12.01	54.03	3.46	19.87	1.01	1.1	1.06	39.9	30.0	9.9
G3	HP	70	7.8	15	470	345.3	10.36	2.07	98.32	10.46	109.8	0.41	12.2	2.2	7.35	3.28	22.99	2.24	1.68	1.42	62.4	41.2	21.2
G4	HP	85	7.9	8	430	315.9	12.66	1.61	67.41	10.24	158.6	0.91	9.66	8.65	7.95	4.45	23.95	1.44	1.44	1.13	58.1	33.7	24.4
G5	HP	65	8	13	470	345.3	12.43	2.46	75.52	10.44	146.4	1.95	33.3	6.94	32.19	3.44	18.04	1.20	1.13	1.16	56.2	40.1	16.1
G6	HP	90	8.1	108	590	433.5	9.44	1.54	95.99	9.61	97.6	0.77	23.4	12.68	8.86	3.01	29.83	1.32	1.01	1.03	42.5	30.4	12.1
G7	HP	60	8.1	22	400	293.9	13.12	2.76	70.59	10.9	140.3	0.74	6.75	2.33	9.04	4.3	14.69	1.00	2.35	1.19	73.2	52.3	20.9
G8	HP	65	8	31	390	286.6	9.44	1.30	93.44	11.11	134.2	0.44	11.8	8.28	7.42	4.82	32.20	1.42	1.46	1.03	76.7	54.8	21.9
G9	TW	130	8.2	82	710	521.7	17.96	1.54	87.29	9.38	97.6	0.94	50	20.23	36.68	3.54	28.83	1.13	0.92	1.06	28.4	20.3	8.1
G10	HP	78	8.1	68	570	418.8	11.97	1.61	101.3	9.95	48.8	1.4	111	26.82	17.52	5.25	24.17	2.09	0.87	1.05	35.1	25.1	10.0
G11	HP	50	8.2	-56	430	315.9	8.75	1.61	98.5	10.93	262.3	0.27	10.4	39.06	13.62	2.58	30.90	0.87	2.81	1.02	194.3	138.8	55.5
G12	HP	45	8.1	-17	550	404.1	10.13	1.92	82.54	11.06	195.2	0.31	20.2	23.44	17.99	2.56	17.50	0.80	2.21	1.06	121.5	71.3	50.2
G13	HP	72	8.2	96	460	338.0	8.75	1.07	79.71	10.09	79.3	0.42	20.6	14.68	11.76	2.52	28.12	0.84	0.85	1.03	36.8	26.3	10.5
G14	OW	40	8	102	490	360.0	12.89	1.84	96.66	10.58	67.1	0.55	29.6	44.29	15.42	2.43	28.33	0.72	0.82	1.04	21.7	15.5	6.2
G15	TW	110	8.2	-54	450	330.6	10.36	2.53	98.21	10.57	85.4	0.44	22	40.5	20.41	2.33	14.15	0.65	1.32	1.47	52.1	37.2	14.9
G16	HP	80	7.8	-110	370	271.9	6.68	2.53	87	11.13	79.3	0.21	12.9	13.62	4.15	2.19	38.48	0.59	1.1	1.43	31.3	22.4	8.9
G17	HP	82	8	55	510	374.7	10.82	2.23	89.63	10.85	122	0.39	10.1	26.09	10.99	2.85	42.16	0.74	1.08	1.26	46.5	33.2	13.3
G18	HP	52	8.3	-78	500	367.4	15.66	1.69	75.75	10.68	67.1	0.43	6.96	34.78	5.28	3.31	32.54	0.90	1.59	1.05	176.6	104.6	72.0
G19	HP	43	8	-54	850	624.6	24.64	3.22	119.3	10.16	54.9	0.47	166	27.52	151.5	2.75	34.83	0.59	1.411	1.05	127.3	75.9	51.4
G20	HP	66	8.2	-25	620	455.6	14.51	1.07	121.8	10.99	128.1	0.33	72.5	36.75	45.55	2.94	27.82	0.67	1.52	1.22	78.3	52.1	26.2
G21	HP	62	8.1	-56	350	275.4	10.36	1.77	59.09	11.08	97.6	0.24	9.59	38.57	9.66	3.35	24.97	0.54	1.47	1.12	68.7	44.6	24.1
G22	TW	95	8.2	92	390	286.6	12.66	1.30	73.42	10.94	128.1	0.55	39.2	26.17	46.47	3.49	32.19	0.84	1.45	1.31	54.2	38.7	15.5
G23	OW	35	8.1	48	360	264.5	8.52	2.23	69.65	11.06	146.4	0.53	9.52	6.59	7.07	2.79	34.33	1.02	1.02	1.06	36.6	26.1	10.5
G24	HP	72	8	47	318	233.7	4.37	1.61	42.5	11.06	115.9	0.67	2.57	1.28	4.72	3.55	30.05	2.32	0.95	1.04	27.4	19.6	7.8
G25	HP	88	8.1	60	400	293.9	13.12	2.76	70.59	10.9	140.3	0.74	6.75	2.33	9.04	3.93	26.15	1.49	0.87	1.19	38.5	27.5	11.0
G26	HP	63	8	-22	400	293.9	7.14	2.76	101.9	11.11	189.1	0.48	11.1	10.56	20.51	3.86	19.78	2.33	1.13	1.41	65.9	47.1	18.8
G27	TW	90	8.1	-34	630	462.9	28.55	1.46	96.09	8.98	134.2	0.78	288	17.65	101.1	3.77	18.77	2.59	1.8	1.03	70.5	48.5	22.0
G28	HP	110	8.1	129	620	455.6	12.89	2.53	98.06	9.93	140.3	0.95	76.1	4.3	55.48	4.68	11.96	2.84	0.87	1.05	26.3	18.8	7.5
G29	HP	100	8.1	52	380	279.2	9.67	1.84	80.58	10.86	73.2	0.67	26.6	10.83	39.57	4.3	30.99	2.74	1.2	1.05	31.4	22.4	9.0
G30	HP	105	8	63	1020	749.5	29.70	5.22	109.2	9.08	146.4	0.53	194	76.77	156.1	4.46	21.44	2.54	1.1	1.1	39.2	28.0	11.2

(HP-Hand Pump, OW- Open Well, TW- Tube Well)

Table 10 Hydro geochemistry of samples in Post-monsoon of Ghazipur (n=30)

ID	Type	Depth (ft)	Ph	ORP mv	EC µs/cm	TDS mg/L	Na mg/L	K mg/L	Ca mg/L	Mg mg/L	HCO <sub>3</sub> <sup>-</sup> mg/L	F <sup>-</sup> mg/L	Cl <sup>-</sup> mg/L	NO <sub>3</sub> <sup>-</sup> mg/L	SO <sub>4</sub> <sup>2-</sup> mg/L	PO <sub>4</sub> <sup>3-</sup> mg/L	SiO <sub>2</sub> mg/L	NH <sub>4</sub> <sup>+</sup> mg/L	Fe mg/L	Mn mg/L	As(tot) µg/L	As <sup>3+</sup> µg/L	As <sup>5+</sup> µg/L
G1	HP	85	8	90	613	435.4	14.10	2.18	86.23	8.85	159.59	0.50	86.72	3.60	47.61	4.01	18.37	0.91	1.20	0.87	35.8	25.5	10.3
G2	TW	120	8.1	95	561	404.3	12.49	1.93	90.12	9.23	144.39	1.04	76.12	9.77	49.88	3.89	22.46	0.85	1.21	0.96	34.5	25.8	8.7
G3	HP	70	7.5	21	421	292.4	9.06	1.68	89.71	10.09	136.79	0.40	10.42	1.79	6.79	3.69	25.98	2.18	1.85	1.29	54.0	35.5	18.5
G4	HP	85	7.5	11	377	267.5	11.08	1.31	61.51	9.88	197.58	0.89	8.26	7.04	7.34	5.01	27.07	1.40	1.59	1.03	52.0	29.0	23.0
G5	HP	65	7.8	17	423	292.4	10.87	2.00	68.91	10.07	182.38	1.90	28.45	5.65	29.72	3.87	20.39	1.17	1.25	1.06	48.6	34.6	14.0
G6	HP	90	8	114	517	367.0	8.26	1.25	87.59	9.27	121.59	0.75	20.03	10.31	6.25	3.38	33.71	1.28	1.11	0.65	37.1	26.1	11.0
G7	HP	60	7.9	25	350	248.8	11.48	2.24	64.41	10.51	174.78	0.72	5.77	1.90	8.35	4.83	16.60	1.10	2.59	1.08	63.3	45.0	18.3
G8	HP	65	7.7	36	342	242.6	8.26	1.06	85.26	10.72	167.18	0.43	10.09	6.74	6.85	5.43	36.39	1.12	1.61	0.94	66.3	47.2	19.1
G9	TW	130	8	92	622	441.6	15.71	1.25	79.65	9.05	121.59	0.92	42.72	16.46	30.24	3.98	26.47	1.10	1.01	0.96	24.6	17.5	7.1
G10	HP	78	7.9	71	499	354.6	10.47	1.31	92.42	9.60	60.79	1.37	95.08	21.82	16.17	5.91	27.31	2.03	0.96	0.96	30.4	21.6	8.8
G11	HP	50	8.1	-41	377	267.5	7.65	1.31	89.88	10.54	326.77	0.26	8.85	31.77	10.24	2.90	34.92	0.85	3.10	0.93	155.4	119.5	35.9
G12	HP	45	7.8	-28	482	342.1	8.86	1.56	75.31	10.67	243.18	0.30	17.22	17.60	16.61	2.88	19.77	0.78	2.44	0.96	87.6	61.4	26.2
G13	HP	72	8	85	402	286.1	7.65	0.87	72.73	9.73	98.79	0.41	17.63	11.94	10.86	2.83	33.28	0.82	0.94	0.94	31.8	22.6	9.2
G14	OW	40	7.6	120	429	304.8	11.28	1.50	88.20	10.21	83.59	0.54	25.33	36.03	14.23	2.74	32.01	0.70	0.90	0.95	18.8	13.3	5.4
G15	TW	110	7.6	-36	394	279.9	9.06	2.06	89.61	10.20	106.39	0.25	18.83	23.10	18.84	2.62	15.99	0.63	1.46	1.20	45.1	32.0	13.0
G16	HP	80	7.4	-76	324	230.2	5.84	2.06	79.38	10.74	98.79	0.30	11.06	11.08	3.83	2.47	43.49	0.58	1.21	1.30	27.1	19.3	7.8
G17	HP	82	7.9	71	447	317.2	9.46	1.81	81.78	10.47	151.99	0.38	8.61	21.22	8.57	3.21	47.65	0.72	1.19	1.15	40.2	28.6	11.6
G18	HP	52	8.1	-43	438	311.0	13.69	1.37	69.12	10.30	83.59	0.42	5.95	28.29	4.87	3.72	30.84	0.87	1.75	0.96	142.6	90.1	52.5
G19	HP	43	7.6	-44	745	528.7	21.55	2.62	108.86	9.80	68.39	0.46	142.13	22.39	139.87	3.10	39.37	0.57	1.56	0.96	110.1	65.4	44.7
G20	HP	66	7.7	-16	543	385.7	12.69	0.87	111.15	10.60	159.59	0.32	61.94	28.40	42.05	3.30	31.44	0.65	1.68	1.11	67.7	44.9	22.8
G21	HP	62	7.8	-52	311	233.2	9.06	1.43	53.92	10.69	121.59	0.23	8.20	31.38	8.92	3.77	25.40	0.53	1.62	1.02	59.4	38.4	21.0
G22	TW	95	7.9	84	342	242.6	11.08	1.06	66.99	10.55	159.59	0.60	33.46	21.29	36.51	3.92	36.38	0.82	1.60	1.19	46.9	33.3	13.5
G23	OW	35	7.8	58	315	223.9	7.45	1.81	63.55	10.67	182.38	0.52	8.14	5.36	6.53	3.13	38.80	0.97	1.12	0.74	31.6	22.5	9.1
G24	HP	72	7.6	61	279	197.8	3.83	1.31	38.78	10.67	144.39	0.65	2.20	1.04	4.36	4.00	33.96	2.01	1.05	0.95	23.7	16.9	6.8
G25	HP	88	7.8	67	350	248.8	11.48	2.24	64.41	10.51	174.78	0.72	5.77	1.90	8.35	4.42	29.55	1.45	0.96	1.08	33.3	23.7	9.6
G26	HP	63	7.7	-31	350	248.8	6.24	2.24	93.01	10.72	235.58	0.47	9.46	8.59	18.93	4.34	22.36	1.50	1.25	1.10	57.0	40.5	16.4
G27	TW	90	7.6	-27	552	391.9	24.97	1.18	87.68	8.66	167.18	0.70	246.57	14.36	93.30	4.24	21.22	2.52	1.98	0.94	61.0	41.8	19.2
G28	HP	110	7.8	112	568	385.7	11.28	2.06	89.48	9.58	174.78	0.93	65.04	3.50	36.47	5.26	13.52	2.77	0.96	0.96	22.7	16.2	6.6
G29	HP	100	7.8	59	325	236.4	8.46	1.50	73.53	10.48	91.19	0.65	22.76	8.81	36.53	4.84	35.02	2.66	1.32	0.75	27.2	19.3	7.8
G30	HP	105	7.6	66	879	634.5	25.98	4.24	99.66	8.76	182.38	0.80	165.49	41.14	84.10	4.14	24.23	2.48	1.21	1.00	33.9	24.1	9.8

(HP-Hand Pump, OW- Open Well, TW- Tube Well)

The depth distribution of arsenic shows strong negative correlation between occurrence of arsenic in groundwater and the depth of hand pumps or tube wells for both seasons shown in fig. (12j & 13j ). In general, the highest arsenic concentration and also the expected spatial and temporal variability, occur 15-25 m below the ground surface, and decreases rapidly below about 40 meters. Similar observations are reported by (Karim et al. 1997) where a maximum arsenic concentration at 20 to 40 m depth, below which arsenic concentration decline. This requires strong redox conditions to drive arsenic mobility along the well depth (McArthur et al. 2001).

From different cross plots shown in fig.(12 d,e,f,g,h,i) and (13 d,e,f,g,h,i) it is clear that the high concentration of arsenic is associated with high concentration of Fe,  $\text{PO}_4^{3-}$  &  $\text{NH}_4^+$  as well as low concentration of Mn,  $\text{NO}_3^-$  and low value of Eh which favors development of anoxic condition during microbial oxidation of sedimentary organic matter along with some anthropogenic output i.e. it might have both the natural and artificial source. Low concentration of nitrate which reveals that it is thermodynamically favourable oxygen source for microbial degradation of dissolved organic materials in the shallow aquifers (HP) of Bhagalpur district. From all the above conditions we can conclude that “Oxyhydroxide reduction theory” is responsible for release of arsenic in aquifer of Ghazipur.

To find the status of the aquifer here we have also taken one core sample in the most effected region of Ghazipur i.e. Devbaranpur upto 110 feet at the interval of 10 feet since at short interval there is no change in lithological composition or granular distribution (shown in fig. 17) and we find that at depth of 30-40 feet, clay percentage is more and it may be responsible for reduced environment.

After speciation it has been observed that As (III) Varies from varied from 15.50  $\mu\text{g/L}$  to 138.78  $\mu\text{g/L}$  in Pre-monsoon and 13.35  $\mu\text{g/L}$  to 119.52  $\mu\text{g/L}$  in Post-monsoon season as shown in (Table15 & 16). The average value of As (III) is equals to 67-69 % of the average value of total As concentration, which is point of attention as it is prime source of lethality. It has been also found that As (III) showing same correlation as by the total As.

#### **Graphical representation of hydrochemical data**

In the present study, water quality data is analyzed through statistical distribution diagrams such as Piper diagram, Durov diagram and Schoeller digram to gain better insight into the hydrochemical processes operating in the groundwater flow system that resulted in the observed spatial and temporal variation in the groundwater quality. Furthermore, the major cations and anions for the analyzed

water were plotted on a Piper diagram to show hydrochemical diversity for Pre-monsoon and Post-monsoon seasons shown in fig. (12 a & 13 a respectively). It has been found that most of the water samples are of Ca-HCO<sub>3</sub> ( about 80 %) and Ca-Cl (about 20 %) type in Pre-monsoon period while in Post-monsoon period most of water samples are also Ca-HCO<sub>3</sub> and Ca-Cl type but in different ratios that are 86.67 % and 13.33 % respectively. This indicates presence of carbonaceous sandstones in the aquifers and weathering of carbonate minerals in Post-monsoon period while in the Pre-monsoon period result favours salt precipitation. In most hardness are of CaCO<sub>3</sub> type. Most of the water samples are saturated with Calcite and Aragonite. From Durov diagram for both Pre-monsoon and Post-monsoon as shown in fig. (12b & 13b) it became clear that most of the water samples are of alkaline in nature and alkalinity decreases in Post-monsoon due to dilution effect. From Schoeller diagram for both Pre-monsoon and Post-monsoon as shown in fig. (12c & 13c) it became clear that Ca is dominant cation while Na & K are very less in value due to poor water- silicate rock interaction. The Post-monsoon samples indicate permanent hardness domain as well as ion exchange and dissolution processes occurring due to recharge of the groundwater in the monsoon period.

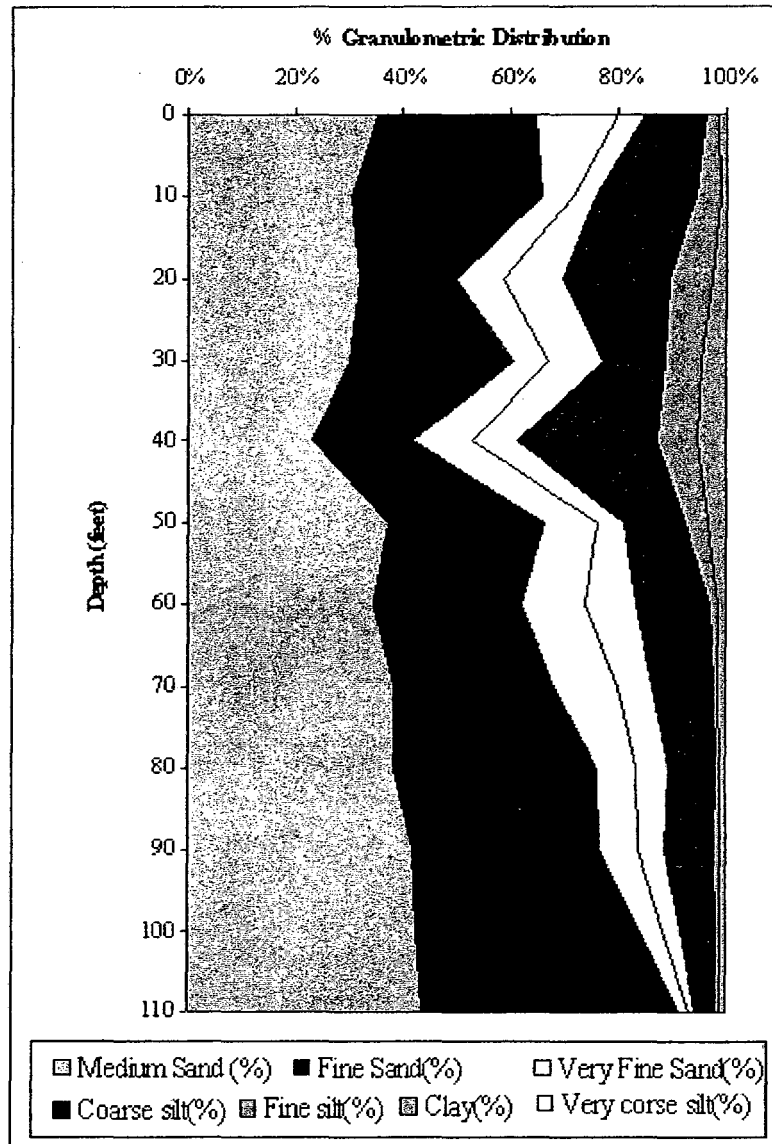


Figure 17- Vertical soil profile of Ghazipur, Uttar Pradesh



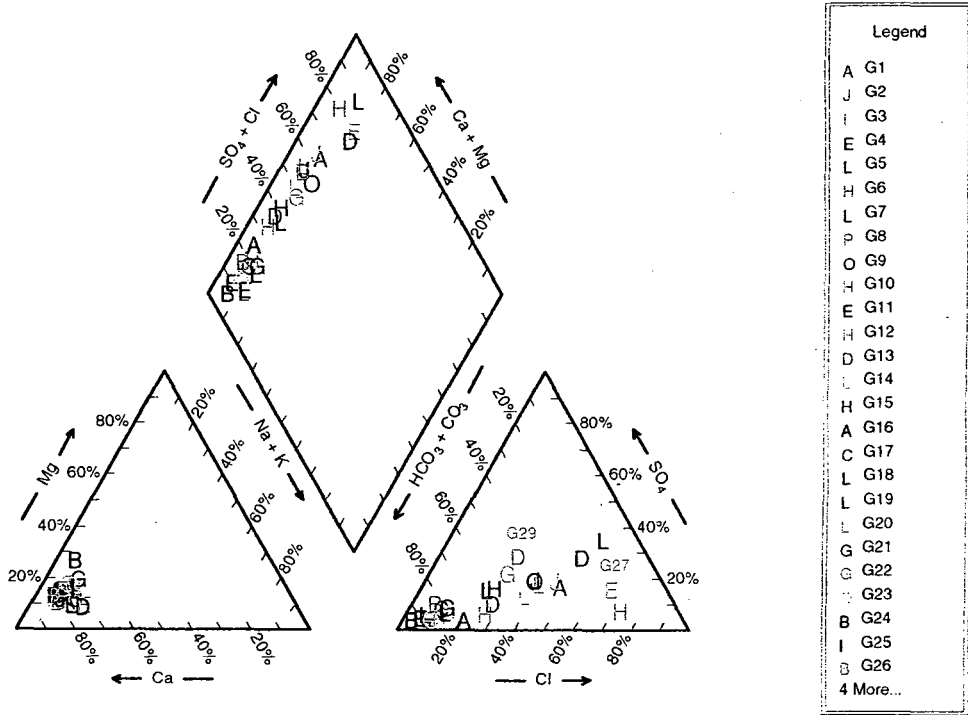


Fig. 12 (a) Piper diagram of Ghazipur for Pre-Monsoon

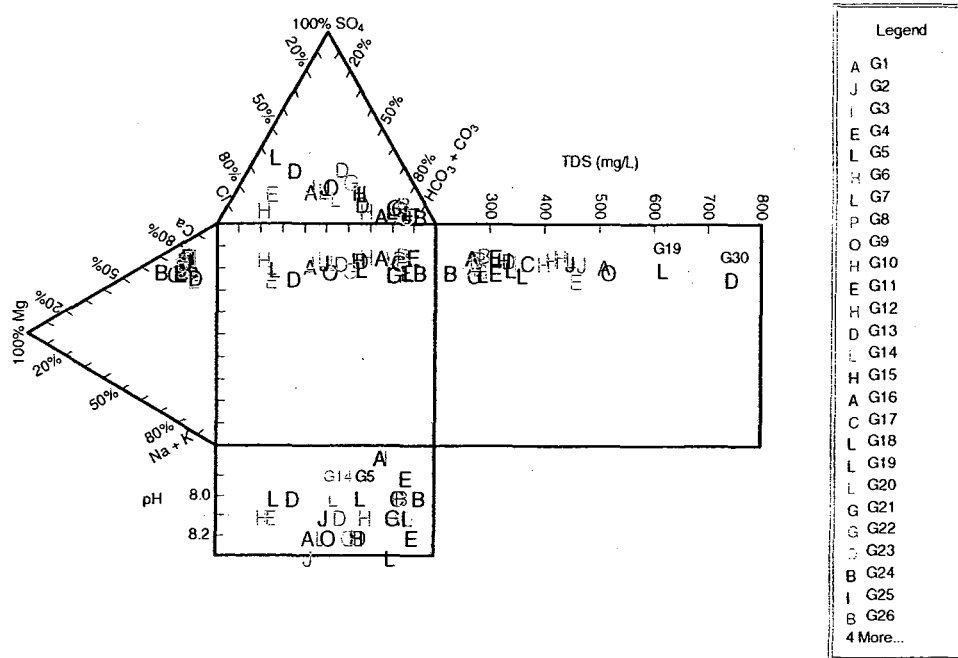


Fig. 12 (b) Durov diagram of Ghazipur for Pre-Monsoon

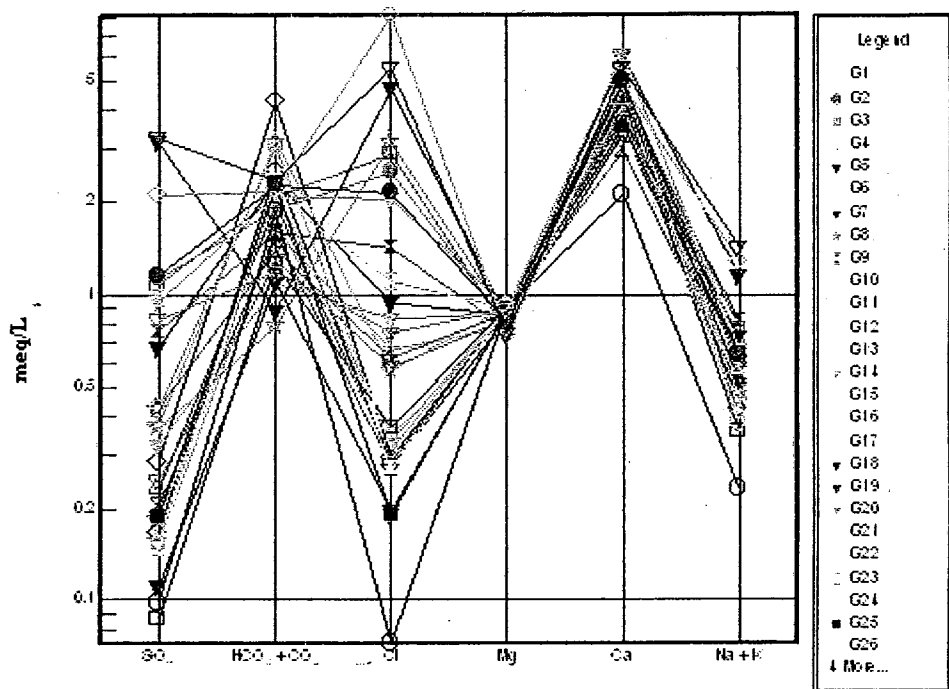


Fig. 12 (c) Schoellar diagram of Ghazipur for Pre-Monsoon

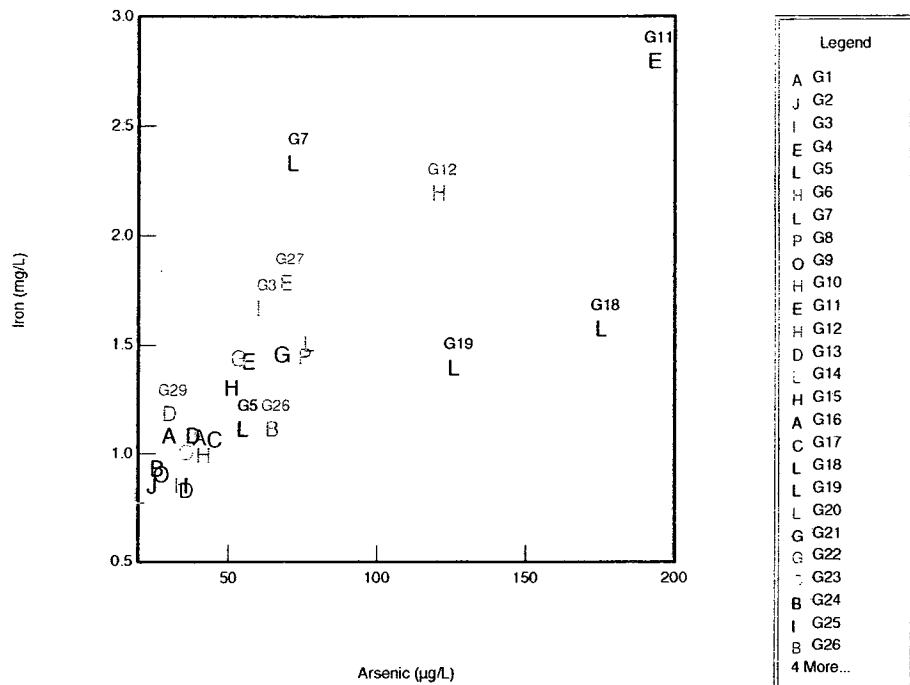


Fig. 12 (d) Scatter diagram of As vs Fe of Ghazipur for Pre-Monsoon

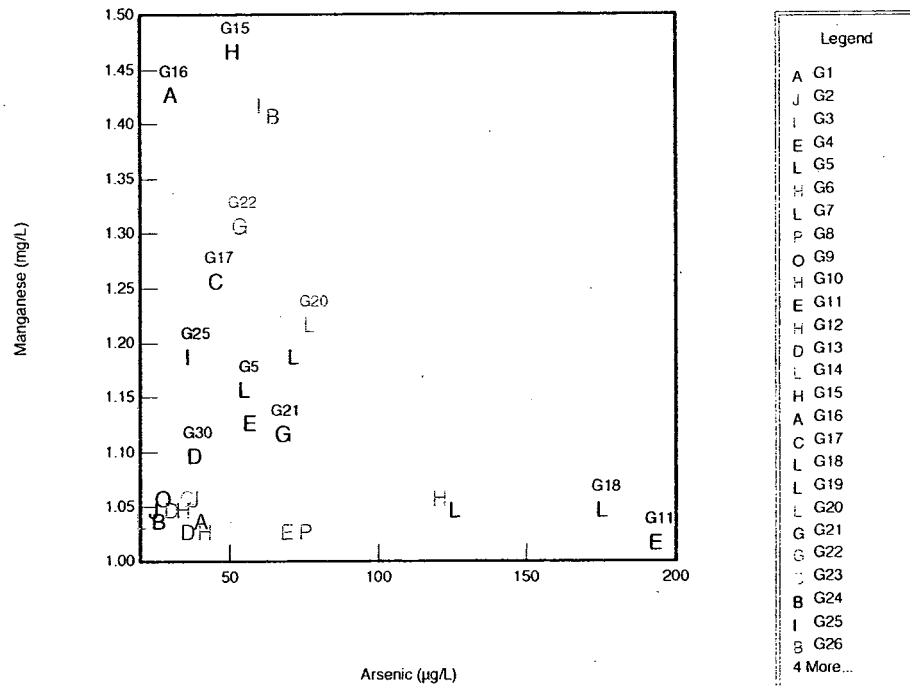


Fig. 12 (e) Scatter diagram of As vs Mn of Ghazipur for Pre-Monsoon

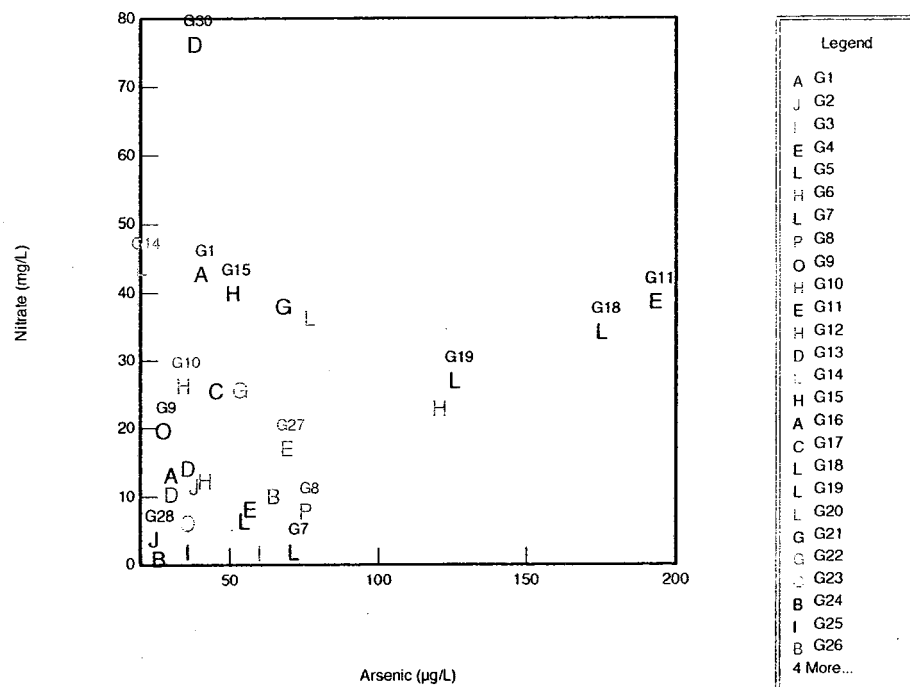


Fig. 12 (f) Scatter diagram of As vs Nitrate of Ghazipur for Pre-Monsoon

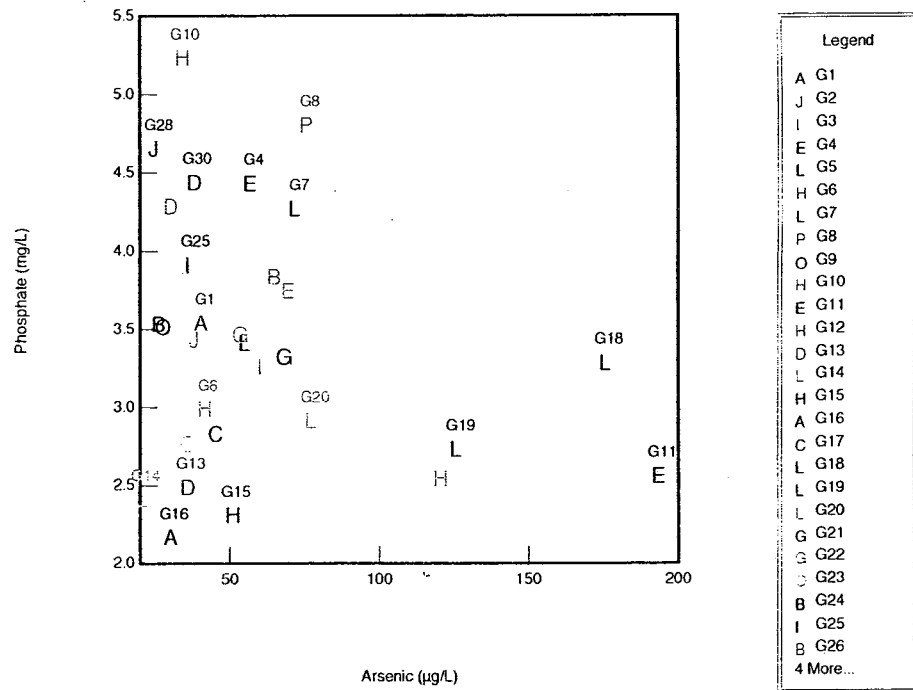


Fig. 12 (g) Scatter diagram of As vs Phosphate of Ghazipur for Pre-Monsoon

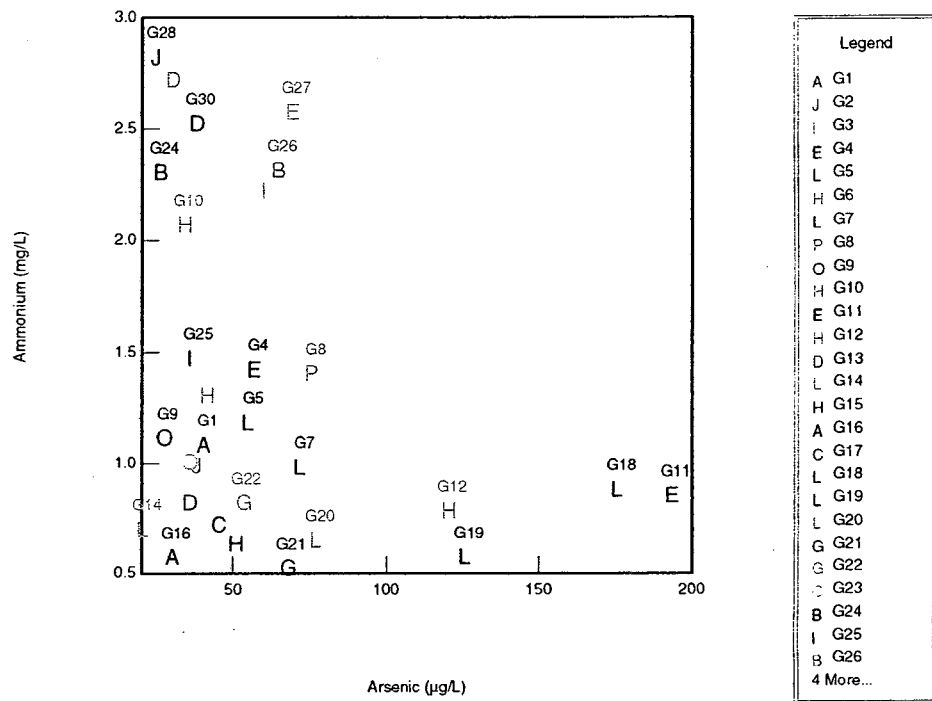


Fig. 12 (h) Scatter diagram of As vs Ammonium of Ghazipur for Pre-Monsoon

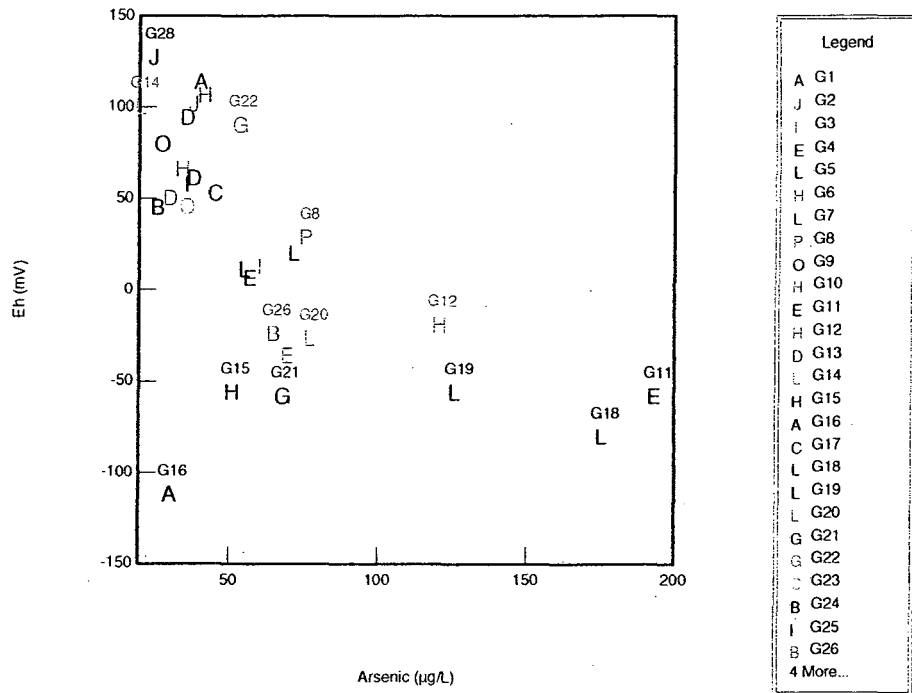


Fig. 12 (i) Scatter diagram of As vs Eh of Ghazipur for Pre-Monsoon

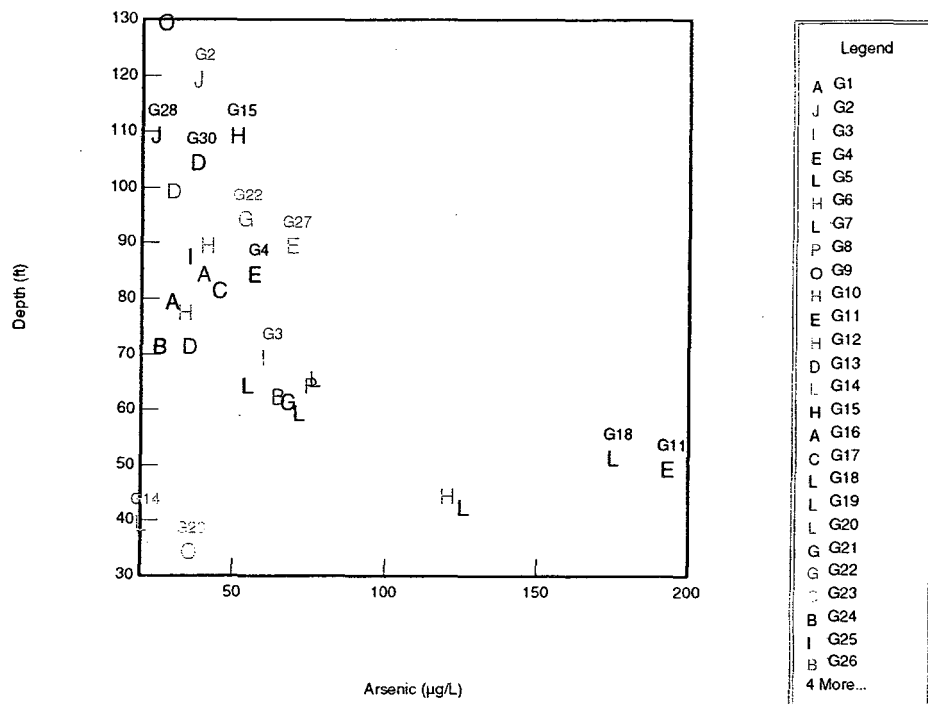


Fig. 12 (j) Scatter diagram of As vs Depth of Ghazipur for Pre-Monsoon

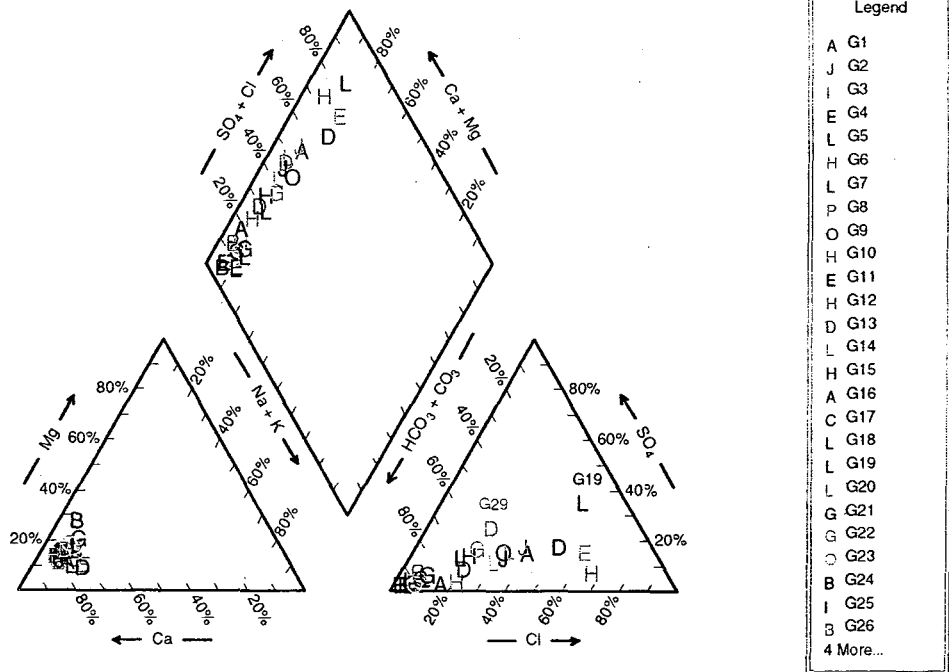


Fig. 13 (a) Piper diagram of Ghazipur for Post-Monsoon

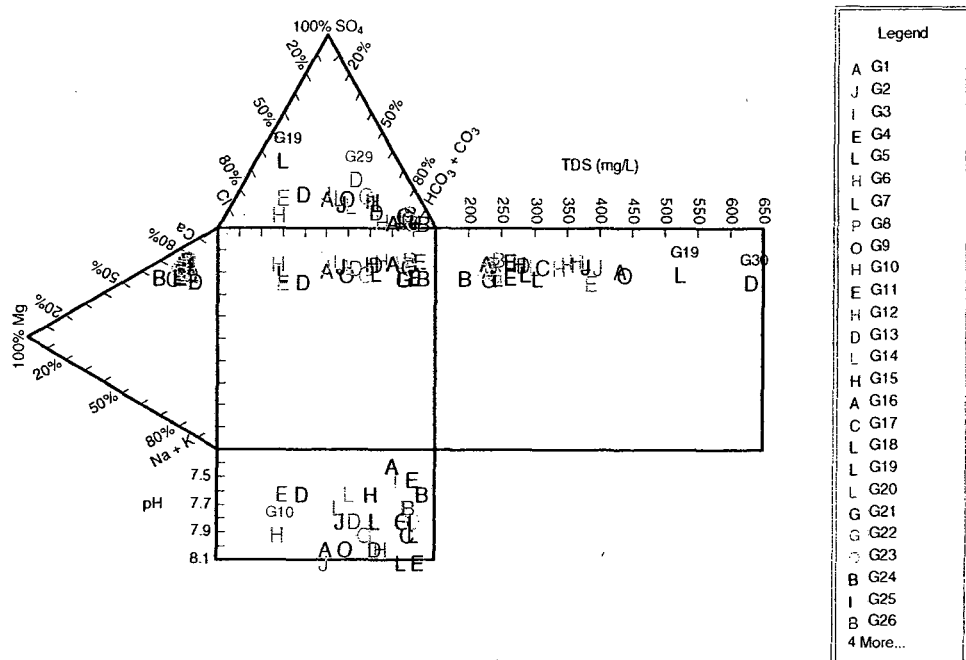


Fig. 13 (b) Durov diagram of Ghazipur for Post-Monsoon

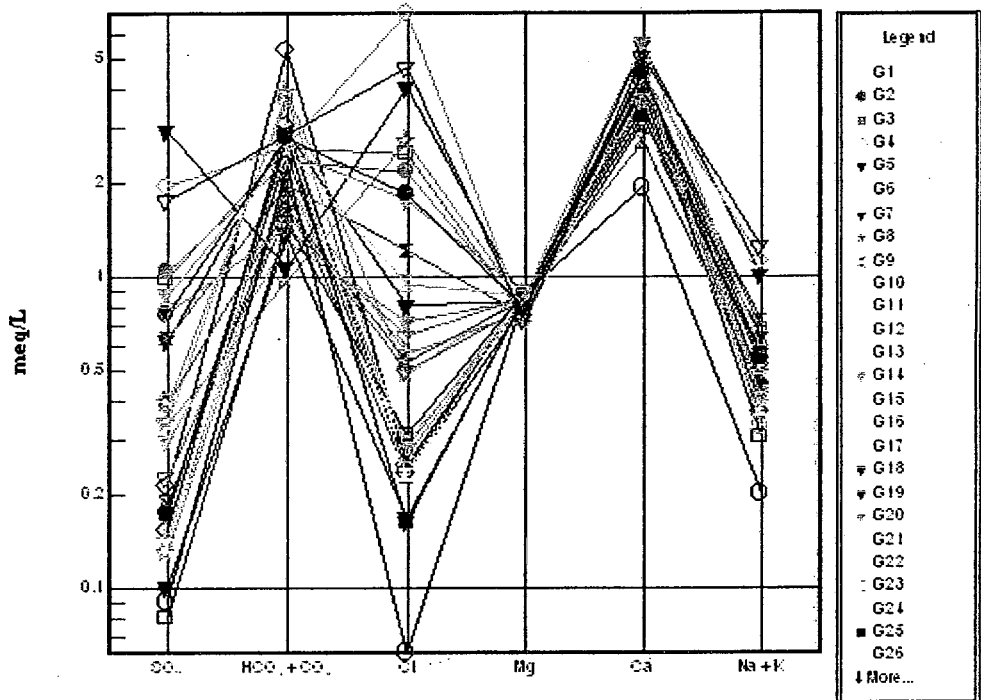


Fig. 13 (c) Schoellar diagram of Ghazipur for Post-Monsoon

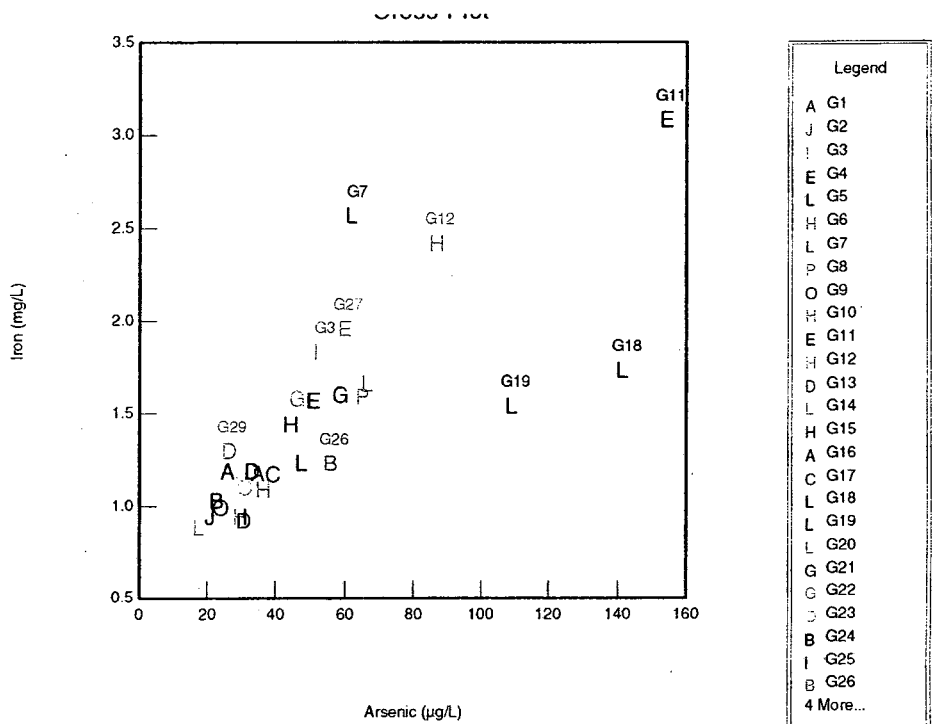


Fig. 13 (d) Scatter diagram of As vs Fe of Ghazipur for Post-Monsoon

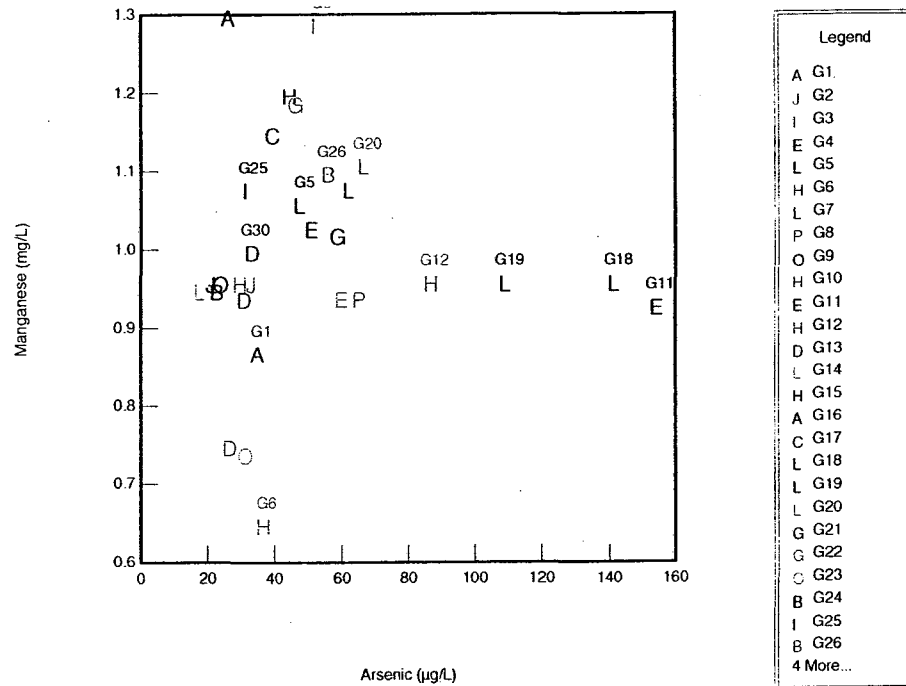


Fig. 13 (e) Scatter diagram of As vs Mn of Ghazipur for Post-Monsoon

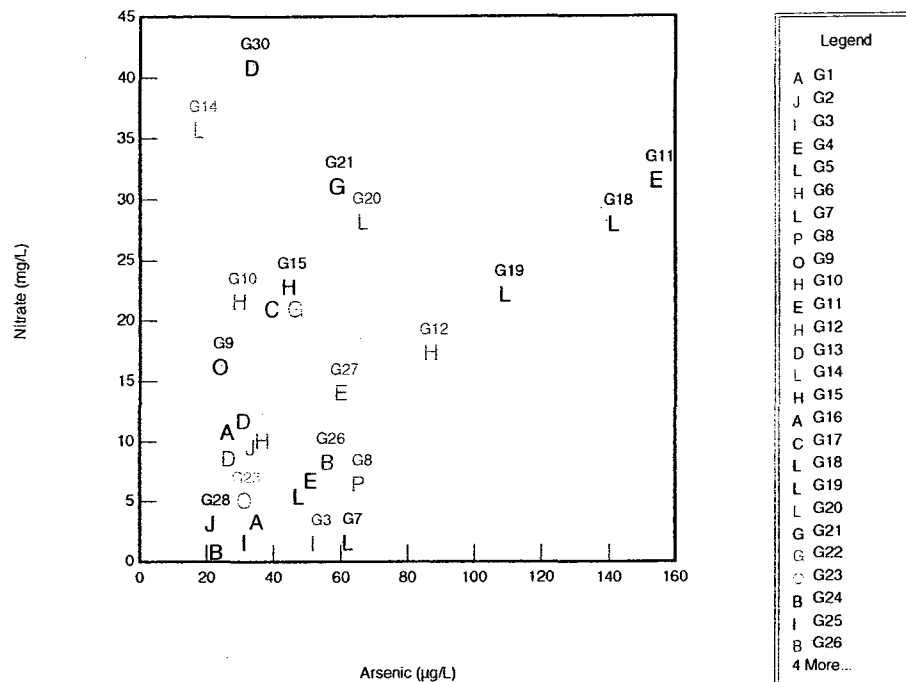


Fig. 13 (f) Scatter diagram of As vs Nitrate of Ghazipur for Post-Monsoon.



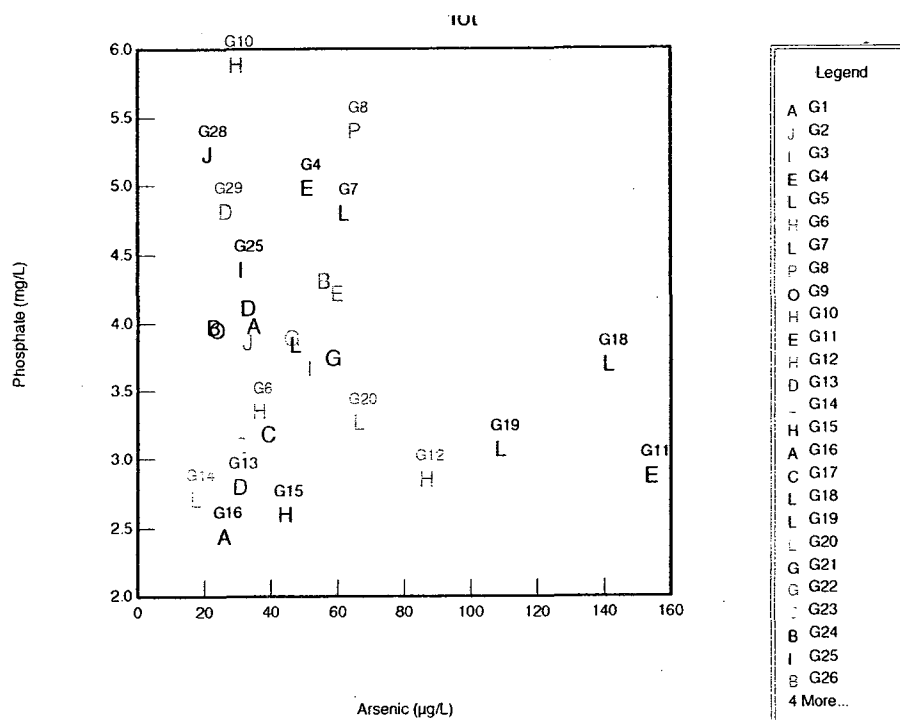


Fig. 13 (g) Scatter diagram of As vs Phosphate of Ghazipur for Post-Monsoon

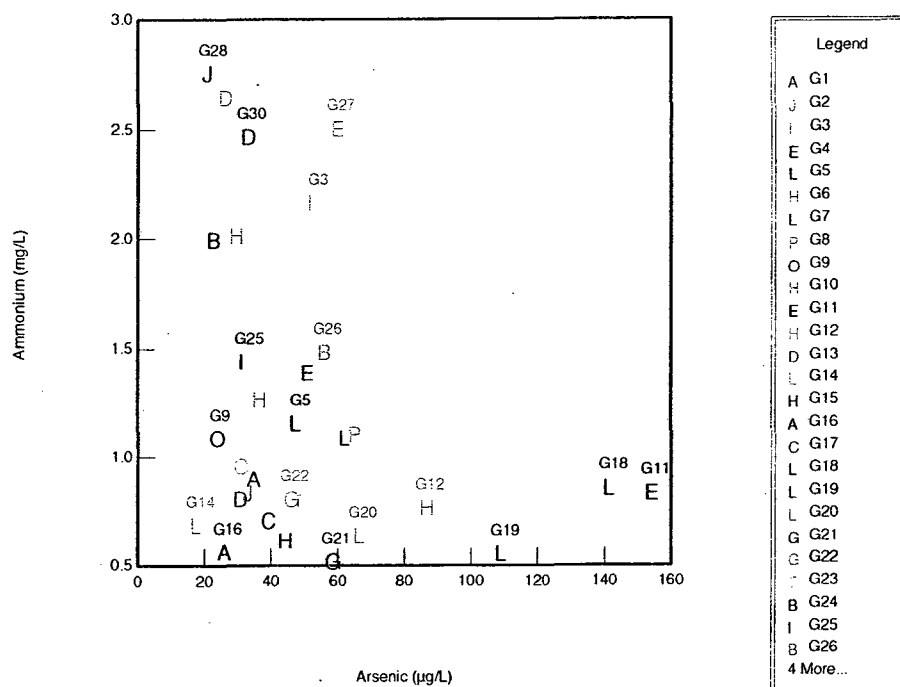


Fig. 13 (h) Scatter diagram of As vs Ammonium of Ghazipur for Post-Monsoon

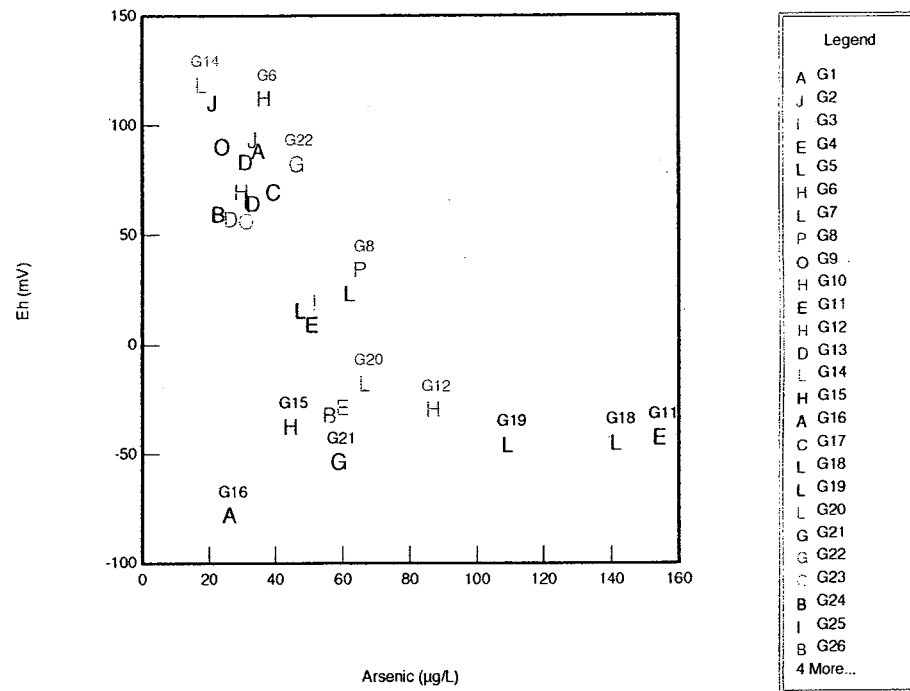


Fig. 13 (i) Scatter diagram of As vs Eh of Ghazipur for Post-Monsoon

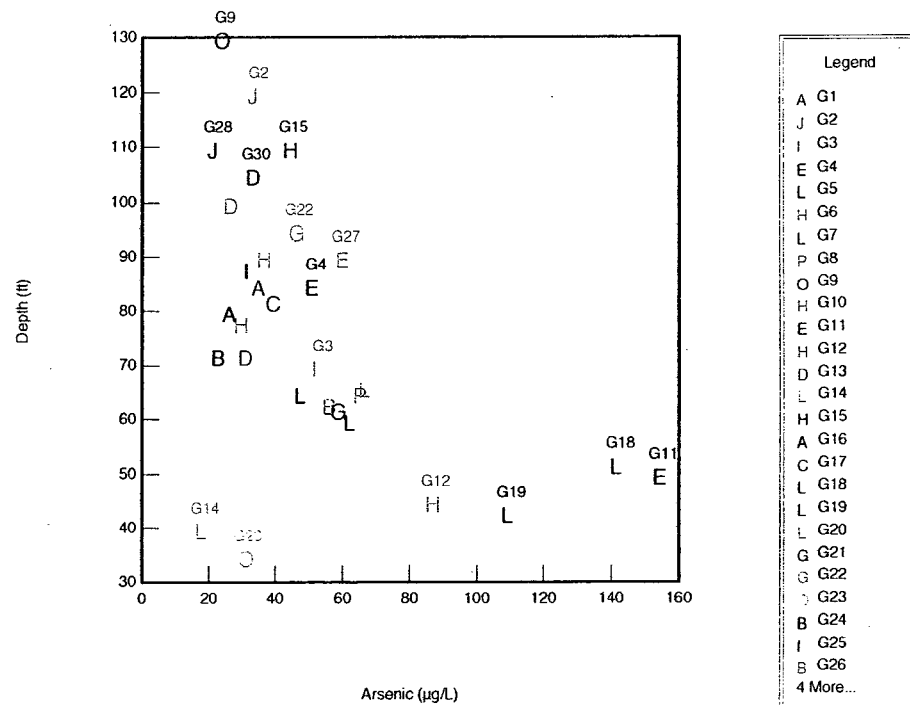


Fig. 13 (j) Scatter diagram of As vs Depth of Ghazipur for Post-Monsoon

### **Statistical analysis**

#### **Correlation analysis**

From table 11 and 12 we find correlation pattern for Pre-monsoon and Post-monsoon respectively for Ghazipur which gives a clear picture of physio-chemical processes occurring there. There was an inverse correlation between As and Eh (Redox potential) i.e. value of Eh increases with the decrease in As concentration, which means concentration of As increases with the increment in reducing condition of this environment. We found a good correlation between As and Fe while an inverse relation between As and ORP as well as with depth which indicates a strong reducing character of this environment. There is a good correlation between Na and  $\text{Cl}^-$  as well as with  $\text{SO}_4^{2-}$  which indicates that the element could be coming from halite or Glauber salt. There are good correlations exist between TDS- Na, TDS- K, TDS - Ca, TDS- $\text{Cl}^-$  & TDS -  $\text{SO}_4^{2-}$  which indicates that carbonate and silicate weathering along with the anthropogenic sources mainly fertilizers and sewage input control the geochemistry of Pre-monsoon ground water. There is also a good correlation found between  $\text{CaSO}_4^{2-}$  &  $\text{Ca-Cl}^-$  which may indicate that both gypsum (absorbed in clay) and limestone are acting as a source of calcium. During post monsoon season most of the correlation is same but intensity became increased.

#### **Correlation related to Arsenic**

The correlation of arsenic and sulphate for Ghazipur is very poor, which indicates arsenic is not released by oxidation of arsenic bearing pyrite or arsenopyrite at rock / water / air interface. Relation between Sulphate with Ca and TDS for both seasons was not so clear to find any promising conclusion as shown in fig.(15), so there is a need to study this area further more with sediment characteristics.

There was an inverse correlation between As and Eh (Redox potential) i.e. value of Eh increases with the decrease in As concentration, which means concentration of As increases with the increment in reducing status of any environment. We found a good correlation of As with Fe,  $\text{NH}_4^+$  &  $\text{PO}_4^{3-}$  While an inverse relation of As with depth, Mn, ORP &  $\text{NO}_3^-$  which indicates a strong reducing character of this environment. This behavior of arsenic is governed by redox reactions at shallow depth which support the dissolution of arsenic from arseniferous iron oxyhydroxides. This assumption is being supported by high correlation of As – Fe.

### **Factor analysis**

Factor analysis of physico-chemical parameters of the Ghazipur groundwater indicates four trends for Pre-monsoon and post- monsoon periods. The total variability accounted for six factors is 80.69 % in Pre-monsoon and while it accounted for five factors is 75.48 % in post- monsoon. The number of significant factors within the data is established by considering only those with an Eigen value >1.0. The degree of association between each variable and each factor is given by its loading on that factor. The Pre-monsoon (Table 13) and post monsoon (Table 14) indicate the loading of each variable on each factor and the communality of each variable accounted for in the analysis.

In Pre-monsoon season, component 1 shows the strong geochemical associations between EC, TDS, Na, K, Ca,  $\text{NO}_3^-$  and  $\text{Cl}^-$  which indicate that amount of weathering as well as anthropogenic activities are mainly responsible for high value of dissolved solid or conductivity. In component 2 there is a strong association between Fe, As, As(III) and As (V) and inverse association with depth and ORP that are indicator of reducing environment which might be responsible for arsenic pollution in the water sample. Component 3 is accounted for  $\text{SiO}_2$  that indicate silicate weathering. Component 4, 5 & 6 accounts for Ph, Mn,  $\text{NH}_4^+$  and  $\text{HCO}_3^-$  which support for anoxic condition as well as for weathering. This indicates the multiple sources controlling the occurrence of As in Ghazipur ground water. In post monsoon season, all components show almost same association but with different intensity.

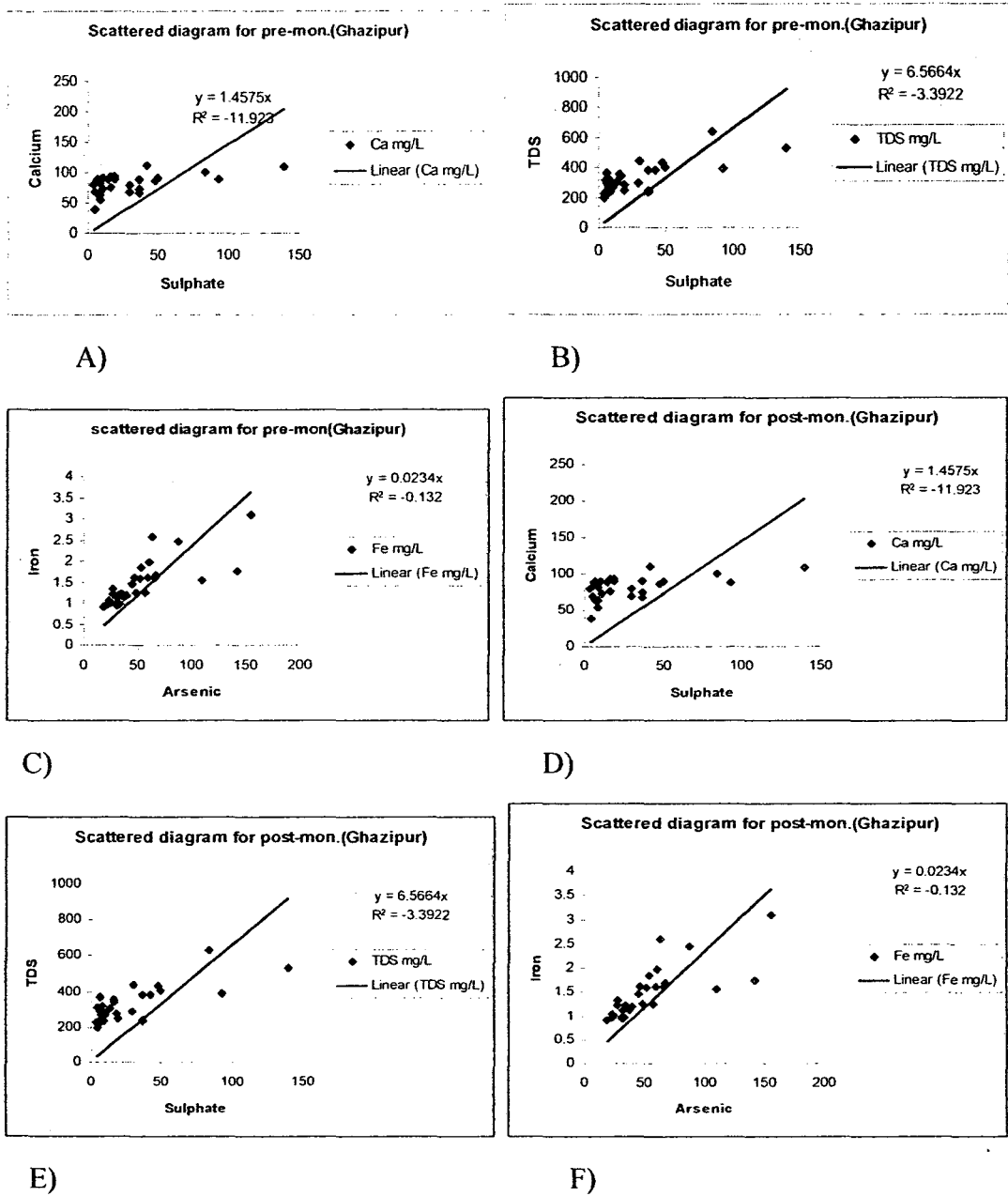


Fig. 15 (A to F) Different scattered diagrams showing relation between Calcium-Sulphate, TDS-Sulphate and Arsenic- Iron for both seasons in Ghazipur.

Table 11 Ghazipur Pre-monsoon correlation matrix (n=30)

	Depth	Ph	ORP	EC	TDS	Na <sup>+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	F <sup>-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	PO <sub>4</sub> <sup>3-</sup>	SiO <sub>2</sub>	NH <sub>4</sub> <sup>+</sup>	Fe	Mn	As(tot)	As <sup>3+</sup>	As <sup>5+</sup>	
Depth	1																						
Ph	0.25	1																					
ORP	0.39	0.22	1																				
EC	0.22	0.28	0.18	1																			
TDS	0.22	0.28	0.17	1.00	1																		
Na <sup>+</sup>	0.13	0.23	-0.07	0.73	0.73	1																	
K <sup>+</sup>	-0.04	-0.23	-0.11	0.16	0.16	0.10	1																
Ca <sup>2+</sup>	0.05	0.07	-0.06	0.67	0.66	0.39	0.13	1															
Mg <sup>2+</sup>	-0.51	-0.27	-0.41	-0.73	-0.73	-0.63	0.02	-0.32	1														
HCO <sub>3</sub> <sup>-</sup>	-0.18	0.03	-0.11	-0.22	-0.22	-0.19	0.02	-0.07	0.23	1													
F <sup>-</sup>	0.32	0.04	0.39	0.20	0.19	0.19	0.11	-0.08	-0.44	-0.08	1												
Cl <sup>-</sup>	0.18	0.15	-0.01	0.68	0.68	0.84	0.03	0.46	-0.68	-0.17	0.23	1											
NO <sub>3</sub> <sup>-</sup>	-0.17	0.42	-0.22	0.29	0.31	0.23	-0.15	0.35	-0.07	-0.14	-0.34	0.16	1										
SO <sub>4</sub> <sup>2-</sup>	0.12	0.15	-0.04	0.73	0.73	0.80	0.26	0.50	-0.49	-0.16	0.13	0.83	0.16	1									
PO <sub>4</sub> <sup>3-</sup>	0.29	-0.03	0.32	-0.04	-0.04	0.10	-0.05	-0.12	-0.17	0.01	0.50	0.15	-0.42	0.0	1								
SiO <sub>2</sub>	-0.24	-0.19	-0.15	-0.17	-0.17	-0.17	-0.27	-0.07	0.35	-0.24	-0.38	-0.23	0.08	-0.1	-0.31	1							
NH <sub>4</sub> <sup>+</sup>	0.30	-0.22	0.26	-0.07	-0.08	0.01	-0.02	-0.04	-0.22	0.06	0.34	0.25	-0.55	0.1	0.65	-0.31	1						
Fe	-0.36	0.09	-0.53	-0.09	-0.08	0.12	-0.08	0.06	0.24	0.57	-0.33	0.01	0.17	0.0	-0.11	-0.14	-0.18	1					
Mn	0.13	-0.38	-0.37	-0.32	-0.32	-0.27	0.32	0.11	0.38	0.01	-0.23	-0.28	-0.05	-0.2	-0.23	-0.04	-0.10	0.01	1				
As(tot)	-0.50	0.24	-0.61	0.10	0.11	0.19	-0.05	0.18	0.22	0.38	-0.33	0.01	0.36	0.1	-0.21	0.10	-0.29	0.77	-0.14	1			
As <sup>3+</sup>	-0.47	0.25	-0.59	0.06	0.06	0.15	-0.06	0.19	0.23	0.45	-0.32	0.00	0.35	0.1	-0.20	0.10	-0.27	0.80	-0.13	0.99	1		
As <sup>5+</sup>	-0.51	0.20	-0.62	0.17	0.17	0.25	-0.03	0.14	0.20	0.25	-0.33	0.03	0.35	0.2	-0.21	0.11	-0.29	0.69	-0.16	0.97	0.92	1	

Table 12 Ghazipur Post-monsoon correlation matrix (n=30)

	Depth	Ph	ORP	EC	TDS	Na <sup>+</sup>	K <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	HCO <sub>3</sub> <sup>-</sup>	F <sup>-</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	PO <sub>4</sub> <sup>3-</sup>	SiO <sub>2</sub>	NH <sub>4</sub> <sup>+</sup>	Fe	Mn	As(tot)	As <sup>3+</sup>	As(5+)	
Depth	1																						
Ph	0.11	1																					
ORP	0.40	0.34	1																				
EC	0.30	0.05	0.18	1																			
TDS	0.30	0.06	0.17	0.91	1																		
Na <sup>+</sup>	0.23	-0.08	-0.01	0.81	0.82	1																	
K <sup>+</sup>	0.13	-0.24	-0.02	0.52	0.52	0.45	1																
Ca <sup>2+</sup>	0.10	-0.07	-0.06	0.66	0.65	0.44	0.25	1															
Mg <sup>2+</sup>	-0.55	-0.17	-0.39	-0.77	-0.77	-0.69	-0.26	-0.38	1														
HCO <sub>3</sub> <sup>-</sup>	-0.15	0.11	-0.18	-0.11	-0.11	-0.10	0.09	-0.04	0.17	1													
F <sup>-</sup>	0.31	0.14	0.40	0.20	0.19	0.18	0.12	-0.08	-0.40	-0.06	1												
Cl <sup>-</sup>	0.25	-0.15	0.00	0.74	0.74	0.87	0.32	0.51	-0.73	-0.11	0.22	1											
NO <sub>3</sub> <sup>-</sup>	-0.14	0.05	-0.22	0.36	0.38	0.37	0.11	0.37	-0.05	-0.09	-0.27	0.25	1										
SO <sub>4</sub> <sup>2-</sup>	0.15	-0.16	-0.08	0.75	0.75	0.83	0.42	0.54	-0.54	-0.13	0.12	0.85	0.24	1									
PO <sub>4</sub> <sup>3-</sup>	0.30	0.05	0.29	0.01	0.00	0.11	0.01	-0.11	-0.18	0.02	0.51	0.16	-0.35	0.01	1								
SiO <sub>2</sub>	-0.28	-0.05	-0.01	-0.24	-0.22	-0.23	-0.24	-0.06	0.38	-0.21	-0.32	-0.23	0.16	-0.11	-0.32	1							
NH <sub>4</sub> <sup>+</sup>	0.37	-0.22	0.28	0.18	0.17	0.24	0.20	0.02	-0.36	0.04	0.37	0.38	-0.28	0.18	0.62	-0.30	1						
Fe	-0.37	0.10	-0.57	-0.12	-0.12	0.05	-0.12	0.03	0.26	0.56	-0.33	-0.03	0.17	0.00	-0.11	-0.14	-0.16	1					
Mn	0.09	-0.43	-0.39	-0.16	-0.16	-0.07	0.15	0.06	0.31	0.02	-0.16	-0.14	0.06	-0.11	-0.16	-0.04	-0.19	0.13	1				
As(tot)	-0.50	0.24	-0.64	0.03	0.03	0.13	-0.10	0.16	0.23	0.35	-0.33	-0.01	0.35	0.12	-0.20	0.07	-0.31	0.77	0.01	1			
As <sup>3+</sup>	-0.48	0.29	-0.61	-0.01	-0.01	0.07	-0.11	0.16	0.25	0.44	-0.33	-0.04	0.35	0.07	-0.21	0.06	-0.30	0.80	0.00	0.99	1		
As <sup>5+</sup>	-0.50	0.11	-0.65	0.10	0.10	0.23	-0.07	0.15	0.18	0.13	-0.31	0.05	0.32	0.22	-0.16	0.09	-0.30	0.63	0.03	0.94	0.88	1	

Table 13- Factor analysis for Ghazipur Pre-monsoon samples (n=30)

Variables	Comp.1	Comp.2	Comp.3	Comp.4	Comp.5	Comp.6	Factor	Eigen Value	Percentage of variance
Depth	0.27	<b>-0.523</b>	0.304	0.431	-0.308		1	6.098	27.719
Ph		0.132	-0.144	<b>0.85</b>	0.248		2	4.605	20.93
ORP		<b>-0.707</b>	0.189	0.268	0.459	0.185	3	2.653	12.059
EC	<b>0.951</b>				0.128		4	1.616	7.344
TDS	<b>0.952</b>				0.129		5	1.445	6.57
Na <sup>+</sup>	<b>0.898</b>	0.113	0.177				6	1.336	6.073
K <sup>+</sup>	<b>0.569</b>	-0.177		-0.4	-0.158	0.471			
Ca <sup>2+</sup>	<b>0.682</b>		-0.138		-0.23				
Mg <sup>2+</sup>	<b>-0.728</b>	0.276	-0.305	-0.34	-0.123				
HCO <sub>3</sub> <sup>-</sup>	-0.144	0.387	0.106			<b>0.781</b>			
F <sup>-</sup>		-0.273	<b>0.649</b>	0.241	0.108				
Cl <sup>-</sup>	<b>0.848</b>		0.311			-0.141			
NO <sub>3</sub> <sup>-</sup>	<b>0.623</b>		-0.496			0.173			
SO <sub>4</sub> <sup>2-</sup>	<b>0.912</b>		0.116	-0.121					
PO <sub>4</sub> <sup>3-</sup>		-0.133	<b>0.795</b>		0.158	0.114			
SiO <sub>2</sub>	-0.188		-0.463	-0.368	0.276	<b>-0.504</b>			
NH <sub>4</sub> <sup>+</sup>	0.154	-0.195	<b>0.77</b>	-0.242		0.148			
Fe		<b>0.846</b>			-0.108	0.292			
Mn	-0.184		-0.2	-0.208	<b>-0.889</b>				
As(tot)		<b>0.955</b>	-0.178						
As <sup>3+</sup>		<b>0.945</b>	-0.171			0.115			
As <sup>5+</sup>		<b>0.923</b>	-0.18		0.108				



Table 14- Factor analysis for Ghazipur Post-monsoon samples (n=30)

Variables	Comp.1	Comp.2	Comp.3	Comp.4	Comp.5	Factor	Eigen Value	Percentage of variance	Cumulative percentage
Depth	0.278	<b>-0.597</b>	0.185		0.32	1	5.894	26.793	26.793
Ph			-0.132	<b>0.887</b>	0.234	2	4.678	21.265	48.058
ORP		<b>-0.709</b>	0.164	<b>0.522</b>		3	2.53	11.501	59.56
EC	<b>0.95</b>			0.126		4	1.986	9.029	68.589
TDS	<b>0.953</b>			0.128		5	1.516	6.89	75.479
Na <sup>+</sup>	<b>0.909</b>	0.108	0.161						
K <sup>+</sup>	<b>0.513</b>	-0.151		-0.363	0.31				
Ca <sup>2+</sup>	<b>0.685</b>		-0.239						
Mg <sup>2+</sup>	<b>-0.754</b>	0.314	-0.236	-0.306	-0.128				
HCO <sub>3</sub> <sup>-</sup>	-0.158	0.386			<b>0.676</b>				
F <sup>-</sup>	0.152	-0.328	<b>0.561</b>	0.23	0.143				
Cl <sup>-</sup>	<b>0.869</b>		0.29						
NO <sub>3</sub> <sup>-</sup>	0.434	0.251	<b>-0.528</b>		-0.126				
SO <sub>4</sub> <sup>2-</sup>	<b>0.866</b>	0.125	0.126	-0.11	-0.151				
PO <sub>4</sub> <sup>3-</sup>			<b>0.832</b>	0.107					
SiO <sub>2</sub>	-0.213		-0.364		<b>-0.668</b>				
NH <sub>4</sub> <sup>+</sup>	0.203	-0.191	<b>0.791</b>						
Fe		<b>0.824</b>			0.358				
Mn	-0.104		-0.264	<b>-0.724</b>	0.277				
As(tot)		<b>0.951</b>	-0.18	0.123					
As <sup>3+</sup>		<b>0.935</b>	-0.198	0.159	0.154				
As <sup>5+</sup>	0.156	<b>0.905</b>	-0.128		-0.107				

The groundwater quality of the Bhagalpur and Ghazipur has a primary problem of arsenic followed by nitrate contamination which needs special attention. The geochemistry of groundwater from pre-monsoon and post-monsoon seasons shows the changes in ionic strength of water. The average ratio trend of  $(Ca^{2+}+Mg^{2+}) / (Na^{+}+K^{+})$  in the pre-monsoon and in the post-monsoon indicate the dominance of carbonate rock weathering in Bhagalpur as well as in Ghazipur ground water. High nitrate concentrations in groundwater of Bhagalpur indicate high intensive agriculture practices or/and anthropogenic activities. The fluoride concentration is high in some location of Bhagalpur. Since the fluoride concentrations are coinciding with high nitrate concentration which shows more anthropogenic than natural input.

The heterogeneous distribution of arsenic in aquifers around the Bhagalpur district show high arsenic concentrations in Rannuchak , Marwa , Sabour , Mahesitilakpur and Sultanganj while in Ghazipur district it is high in Devbaranpur, Jamania, Gahmar, Dildar Nagar, Fulli, Karanda, Mohammadabad and Reotipur. It has been found that the generally concentration of arsenic is more near to the river in shallow aquifers or where high meandering of river takes place. This behavior of arsenic is governed by redox reactions which support either the dissolution of arsenic from arseniferous iron oxyhydroxides. For Bhagalpur, good correlation ( $>0.5$ ) between Sulphate with Ca and TDS indicates that the acid produced from oxidation of pyrites is being neutralized by carbonate formed during weathering process, and arsenic released consequently possibly by sorbed in hydrated Fe-oxide also produced during weathering. High correlation between arsenic and phosphate shows the mobilization of arsenic may partially governed by anthropogenic agricultural activities. This assumption is being supported by high correlation of As – Fe. For Ghazipur, since results are not so easily predictable so cant conclude much-more about the chemical processes occurring their.

Arsenic speciation shows that the proportion of arsenite ( $As^{+3}$ ) relative to arsenate ( $As^{+5}$ ) in aqueous samples are more. This indicate the shifting behaviour of river Ganga which may lead to variation in hydrological budget which further trigger the reduction of oxidized As to reduced As, a more mobile and toxic species. The Fe – As relation in redox atmosphere is also being proved by paleo-channels and shifting behavior of river Ganga. Bhagalpur district also experiences frequent floods every year during south-west monsoon and is having channelized natural water system, so the ground water frequently interact with the surface water, which accelerate the

weathering and other hydro geochemical processes and consequently trigger the sediment load and accumulation of arsenic in these aquifers. Occurrence of high concentrations of arsenic in down stream of river Ganga is due to the enrichment from transportation of arsenic through river sediments load and these arsenic rich sediments are being deposited in these flood prone regions.

Furthermore, this study needs additional geochemical investigation of surface sediments and core sediment to exactly assess the mineralogical control over the arsenic mobility at different depth in groundwater of central Gangetic plain. But nevertheless the study interdentally shows the arsenic prone regions and its natural and anthropogenic control over its distribution and mobilization in the groundwater of the two districts i.e. Bhagalpur (Bihar) and Ghazipur (Uttar Pradesh) in central Gangetic plain.

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Sample no.	latitude	longitude	Loc. Name
1	86.9457778	25.25749074	Alamgirpur 1
2	86.9444168	25.25476881	Alamgirpur 2
3	86.8110431	25.24524208	Mahesi Tilakpur 1
4	86.8110431	25.24388112	Mahesi Tilakpur 2
5	86.7429953	25.24660304	Sultanganj
6	86.7076106	25.2248278	Masdi
7	86.7661317	25.24660304	Sultanganj Thana
8	86.8695642	25.23843727	Bhavnathpur
9	86.876369	25.24524208	Adarsh Nagar
10	86.8804519	25.2588517	Rannuchak
11	86.8709252	25.21257914	Golgamma
12	86.8382622	25.18399908	Panchrukhi 1
13	86.8382622	25.18127716	Panchrukhi 2
14	86.8396232	25.18127716	Panchrukhi 3
15	86.8423451	25.1526971	Shahkund 1
16	86.8423451	25.15133614	Shahkund 2
17	86.8573157	25.24115919	Akbar Nagar
18	86.937612	25.17447234	Shadhuddin Chawk
19	86.9770798	25.20713529	Aliganj
20	86.9716359	25.19624774	Makhana
21	86.9906892	25.17583331	Puraini
22	86.9784407	25.15814095	Jagdishpur 1
23	86.9784407	25.15541902	Jagdishpur 2
24	87.1199801	25.41672241	Naughachhia 1
25	87.121341	25.4208053	Naughachhia 2
26	87.0342399	25.41944434	Khari
27	86.9580263	25.41263968	Bihpur
28	86.9457778	25.42352723	Marwa
29	86.8940614	25.40855679	Narayanpur
30	87.0669028	25.24388112	Sabour
31	87.0914	25.2248278	Ghoga Bazar
32	87.2234127	25.20169159	Ekchari
33	87.2397441	25.2751831	Kahalgaon 1
34	87.2397441	25.27246117	Kahalgaon 2
35	87.3499816	25.24115919	Shiv Narayanpur
36	87.4425266	25.34050898	Pirpanti

Table (i)- Sampling locations with their latitude &amp; longitude for Bhagalpur, Bihar



Sample no.	latitude	longitude	Loc. Name
1	83.80835806	25.42080729	Saidpur 1
2	83.80923396	25.41993139	Saidpur 2
3	83.81186165	25.44971168	Raghuvar Ganj
4	83.80923396	25.47073311	Tajpur Dehma
5	83.80397858	25.49087863	Rajapur
6	83.7987232	25.52416251	Devkali block
7	83.80397858	25.57233664	Mohammadabad
8	83.79084022	25.5837232	Karanda
9	83.83200717	25.59773754	Ghazipur city 1
10	83.83113128	25.59773754	Ghazipur city 2
11	83.79609551	25.63452501	Devbaranpur
12	83.76281163	25.6380286	Dildar Nagar
13	83.73653482	25.63890449	Dandimorh 1
14	83.73741072	25.6380286	Dandimorh 2
15	83.81624103	25.66693299	KotiyoRadhopur
16	83.78646074	25.68182314	Nawali
17	83.78646074	25.68182314	Tarighat
18	83.81010985	25.70196867	Jamania
19	83.79346791	25.71423122	Gahmar
20	83.7706947	25.72386599	Fulli
21	83.7987232	25.75101869	Bara 1
22	83.7978473	25.75101869	Bara 2
23	83.85390449	25.63452501	Shahbazkuli
24	83.86266336	25.67306427	Gauspur
25	83.8530286	25.72561778	Mohammadabad
26	83.8258759	25.72649368	Reotipur 1
27	83.82850359	25.78079898	Reotipur 2
28	83.88193299	25.71160353	Sukhdehra
29	83.89419545	25.73525264	Qasimabad
30	83.88456068	25.7737919	Karimuddinpur

Table (ii)-Sampling locations with their latitude &amp; longitude for Ghazipur, Uttar Pradesh