

AL. Ramanathan  
Scott Johnston  
Abhijit Mukherjee  
Bibhash Nath *Editors*

# Safe and Sustainable Use of Arsenic- Contaminated Aquifers in the Gangetic Plain

A Multidisciplinary Approach

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A Multidisciplinary Approach

 Springer



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# Foreword

There is a lack of detailed understanding on the primary and secondary sources controlling the spatial variability of arsenic in alluvial aquifers in the Gangetic plain. Further the factors controlling the temporal changes in arsenic concentrations are of great concern now since arsenic toxicity creates an impediment to water usage for agriculture and drinking water purposes. These knowledge gaps critically affect our scientific understanding and ability to develop informed policy to mitigate and manage arsenic contamination. The book thus addresses the extent to which the reductive dissolution of As-bearing Fe(III) oxides hypothesis is applicable, sustainability of the deeper aquifer system(s) as an alternate arsenic-free water source, vertical connectivity between the shallow and deep aquifer system(s) and the lateral heterogeneity of aquitards/semi-confining layers, type of groundwater extraction strategies required to maintain the sustainability of the deeper aquifer system(s) and institutional approach to address and mitigate arsenic issues. In order to resolve these extant knowledge gaps, this book has been brought out as a research frontier to address this public health problem faced by millions of people, through highly integrated, multi-disciplinary hydrogeochemical research. Further it attempts to integrate quantification of arsenic mobilization, sustainable abstraction of arsenic-safe drinking water, predication of the long-term viability of As-safe groundwater, quantifying reactive transport processes of arsenic and simultaneous and comprehensive documentation of As-contaminated shallow aquifer systems.

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# Preface

The Arsenic problem in the groundwater of the Gangetic fluvial plains has direct consequences for human health and is presumed to be one of the biggest natural groundwater calamities that mankind is facing. The Gangetic basin is affected by arsenic contamination in the groundwater that is above the permissible limit of 10  $\mu\text{g/L}$ . People are chronically being exposed to drinking arsenic-contaminated hand tube-wells water from Holocene aquifers (recent alluvial sediments). The arsenic distribution in this region has routes originating from the Himalayan region, and its ingestion through the food chain has far-reaching consequences including health hazards and socio-economic impacts. The rapid spreading of arsenic toxicity in these ground waters is due to the slow to moderate movement and over-exploitation.

Despite taking a number of precautionary measures by various agencies, the arsenic contamination in ground water continues to be an unsolved social problem. To resolve this problem in an adequate manner it desperately needs the bonding of strategic scientific research. This book thus addresses numerous scientific investigations in the Central Gangetic plain and comes out with a number of findings, and alternative propositions, which are collectively presented in this book reflecting exclusively the authors' view and diverse perspectives from their intensive research outputs. The chapters are thus aimed at highlighting the state-of-affairs of arsenic in ground water with scientific narration done in different states. These chapters also discuss the knowledge gaps and areas in which further actions are to be taken up along with their scope.

Section I covers chapters on the role of fluvial geomorphology, quaternary stratigraphy and sedimentology in arsenic distribution using hydrogeochemical evolution and resistivity survey approaches in the different shallow aquifer of Central Gangetic plain. Section II has four chapters covering groundwater topography and aquifer characterization and risk assessment in public groundwater wells along with the existing challenges in managing arsenic contamination.

The chapters in Section III deal with arsenic mobilization and distribution processes and their temporal and spatial variations. Spatio-vertical heterogeneity and surface generated organic matter act as a driver in arsenic mobilization in



these basins including delta and coastal regions are also discussed in detail. Section IV emphasizes “arsenic and health” and its impact on the food chain. This section also encompasses innovative new remediation techniques including the low-cost arsenic removal method.

Thus, this book offers a meaningful and practicable guidance for the better management of arsenic problems in the ground water of the Gangetic plain with contributions from distinguished scientists from both academic and research institutions from all over the world, who have been actively working in this area. We would like to thank all the authors for their contributions and the publishers for bringing out this volume successfully. This book will be of great treasure for those working and planning to work on ground water, hydrogeology, hydrogeochemistry, water quality and other issues related to drinking water quality and the remediation techniques. This book is recommended for all libraries of universities, colleges and other institutions working on water and will be an invaluable reference work for planners who are working on water supply and sanitation.

New Delhi, India  
East Lismore, Australia  
Kharagpur, India  
Sydney, Australia

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**Scott Johnston** an Australian Research Council Future Fellow is based at Southern Cross University, Australia. He has received his Ph.D. in 2005 (UNE) on hydrology and biogeochemistry in sulfidic wetlands. His main work, including more than 45 peer-reviewed manuscripts, stresses on how to unravel the physical and hydrological characteristics of floodplain sedimentary environments. He has worked closely with the Australian industry, state and local governments to develop and refine techniques for remediating degraded floodplain wetlands.

**Abhijit Mukherjee** Assistant Professor, Geology and Geophysics, IIT Kharagpur, India, did his Ph.D. from the University of Kentucky and post-doctorate from University of Texas on hydrogeology and arsenic geochemistry of Indo-Gangetic plain. He is serving as an Associate Editor, *Journal of Hydrology and Applied Geochemistry*. He has done extensive work on elevated arsenic in deeper groundwater of western Bengal basin, India. His work, including more than 30 peer-reviewed articles, also relates to arsenic and other toxic elements in natural water systems, groundwater recharge in natural dune systems, and agricultural ecosystems in the desert region.

**Bibhash Nath** Research Associate at the School of Geosciences, The University of Sydney, is an 'Environmental Geochemist' with specialization in 'Arsenic Hydrogeochemistry'. Dr. Nath researched in Taiwan on water quality issues, especially hydrogeology and geochemistry of arsenic contaminated coastal aquifer in SW Taiwan. He is also a recipient of the prestigious DAAD fellowship (2003).

Dr. Nath has co-authored 52 peer-reviewed journal articles, three book chapters, two technical reports, one editorial and more than 30 conference abstracts/proceedings which have been highly cited. He is an Associate Editor of the *Journal of Hydrology* and a Guest Editor (special issue on ‘Arsenic Ecotoxicology’) of the *Journal of Hazardous Materials*.

**Section I**  
**Role of Fluvial Geomorphology and**  
**Sedimentology in Arsenic Distribution**



# Chapter 1

## Hydrogeochemical Evolution in the Different Shallow Aquifers of Central Gangetic Plain and Kosi Alluvial Fan and Their Implications for the Distribution of Groundwater Arsenic

Abhijit Mukherjee

### 1.1 Introduction

For a long time it was understood that the extent of the arsenic (As) enriched groundwater is confined within the lower Ganges plain and delta in eastern India. However, during the last few years, reports of elevated As in groundwater of different parts of the middle portions of the Gangetic plain, upstream from West Bengal, in U.P. (Ramanathan et al. 2006) and Bihar, got published. Subsequently, As contamination was detected in foothills of Himalayas in Nepal (Shrestha et al. 2003), which is a sediment provenance for many of the tributaries of the Ganges, and also in the Indus alluvial plains in Punjab and Sindh provinces of Pakistan (Nickson et al. 2005). In spite of some surveys on As distribution, there is a lack of hydrogeochemical knowledge about the distribution, extent, severity, source and cause of the contamination in these areas. However, initial estimates show that the poisoning might be widespread and several million people may be at risk.

The groundwater chemistry of the Gangetic aquifers and its associated tributary basin aquifers e.g. the river Kosi fan is suggested to be controlled by the presence of carbonates, the composition of silicates, and the oxidation of sulfides (Galy and France-Lanord 1999). According to these authors, in scarcity of Ca-plagioclase in the Himalayas, weathering of the alkaline Himalayan silicates releases Na and K as the dominant cations. Dissolution of hydrobiotite, vermiculite, and smectite of the sediments derived from the Himalaya (Baumler and Zech 1994; Grout 1995), and introduced from weathering of biotite, are the source of Mg in the groundwater (Galy and France-Lanord 1999). However, Dowling et al. (2003) suggested that the groundwater chemistry is indicative of dissolution of detrital carbonates in the upper

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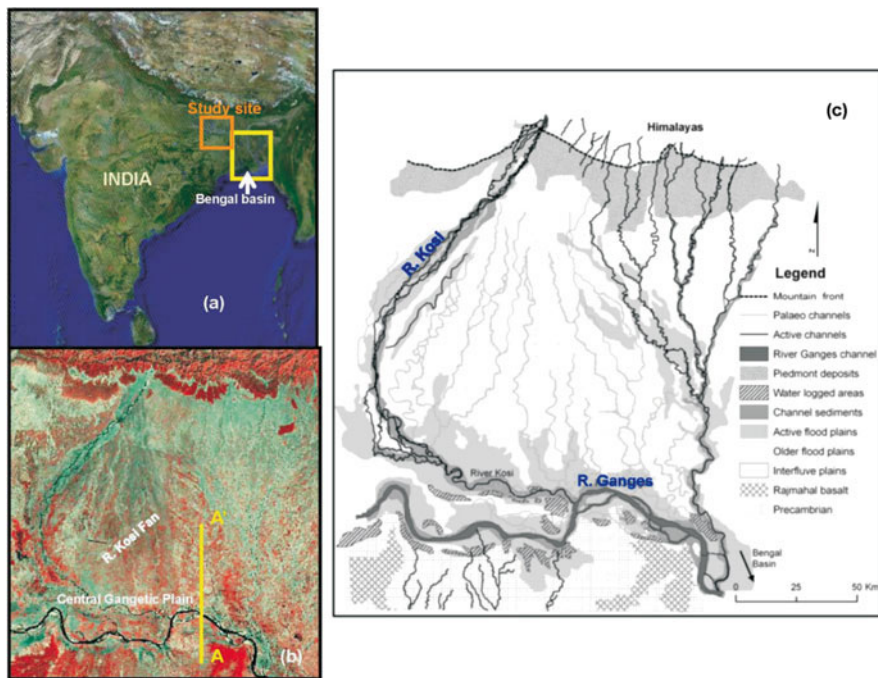
reaches of the river leading to dominance of carbonate weathering. However, proponent of the silicate weathering hypothesis suggested that the chemical evolution would have been dominated by the deposition of silt-dominated sediments in the foreland basin from the rising Himalayas (Burbank 1992; Derry and France-Lanord 1996; Galy and France-Lanord 1999; Kumar et al. 2006). Nevertheless, it is believed that most of the weathering in basin is caused by  $\text{H}_2\text{CO}_3$  liberated by degradation of organic matter in the soil, and <10 % of the weathering is caused by  $\text{H}_2\text{SO}_4$  derived from sulfide oxidation. The groundwater has concentrations of  $\text{NO}_3^-$  (<1–120 mg/L) and  $\text{SO}_4^{2-}$  (15–379 mg/L), suggesting a relatively different redox environment than the Bengal basin groundwater. The observed Eh ranges from 0.12 to 0.7 V, indicating oxic to slightly iron reducing conditions. Such a redox condition might not be conducive of and would limit the microbial reduction of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and Fe(III) (Kumar et al. 2010; Seyler and Martin 1989). A detailed hydrogeochemical characterization of the study area has been provided in Mukherjee et al. (2012).

Shallow groundwater were sampled during the present study along few regional transects, which covered the majority of the aquifers encountered in the central Gangetic plain, comprising the states of Bihar and Jharkhand. The detailed hydrogeology of the area, till date, is largely un-comprehended. The objective of this study is to decipher the regional hydrochemical conditions and trends in various aquifers of the study area, and their influence on the distribution of the groundwater As. The importance of the study is in the fact that the study area is hydrologically and sedimentologically just upstream of Bengal basin (Mukherjee et al. 2009a), and thus the study results can provide immense insight on the conducive environment, conditions and mechanisms for groundwater As mobilization in various geologic-geomorphic terrains, which are obscure in the predominantly fluvio-deltaic Bengal basin.

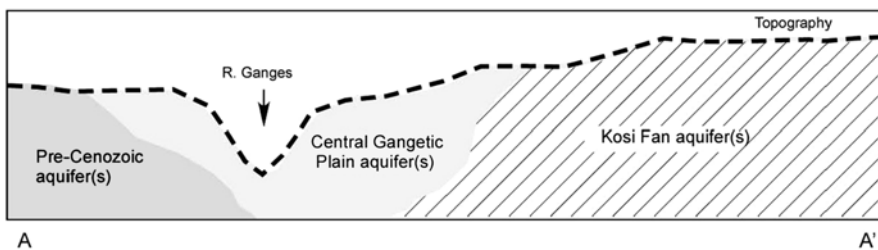
## 1.2 Study Area

The study area extends over the flood plain deposits of the river Ganges (also known as the river Ganga) and its tributaries in the states of Bihar and Jharkhand in eastern parts of India (Fig. 1.1). The area is stretched from Indian cratonic outcrop rocks, representing Peninsular India, and extending up to the Terai plains in north, located in the piedmonts of the Himalayas near the Indo-Nepal international boundary. The southern bank lithology of the river Ganges are composed mostly of Pre-Cenozoic metamorphics, along with some igneous rocks, mostly as volcanics of Rajmahal flood basalt, overlain by a layer of alluvial flood plain deposits. In the northern bank of the river Ganges, the fluvial sedimentation are much more pronounced, and together with the alluvial deposits of river Kosi (or river Kaushiki in Sanskrit), one of the primary tributaries of the Ganges forms the extensive alluvial deposits of the Kosi alluvial fan, which, with its thick coarse to medium grained sand aquifer systems, is sometimes regarded as the “bread basket” for Northern India.

The alluvial deposits in the northern bank of the river Ganges and the Kosi fan is made up of thick pile of unconsolidated Quaternary Alluvium (Fig. 1.2) with several



**Fig. 1.1** Maps of the study area showing (a) disposition of the study area in the Indian-subcontinent, and with reference to the Bengal basin; (b) a FCC (false colour coded) imagery of the study area highlighting the geological and hydrological features; and (c) delineated geomorphologic features within the study area (Modified from Mukherjee et al. 2012)



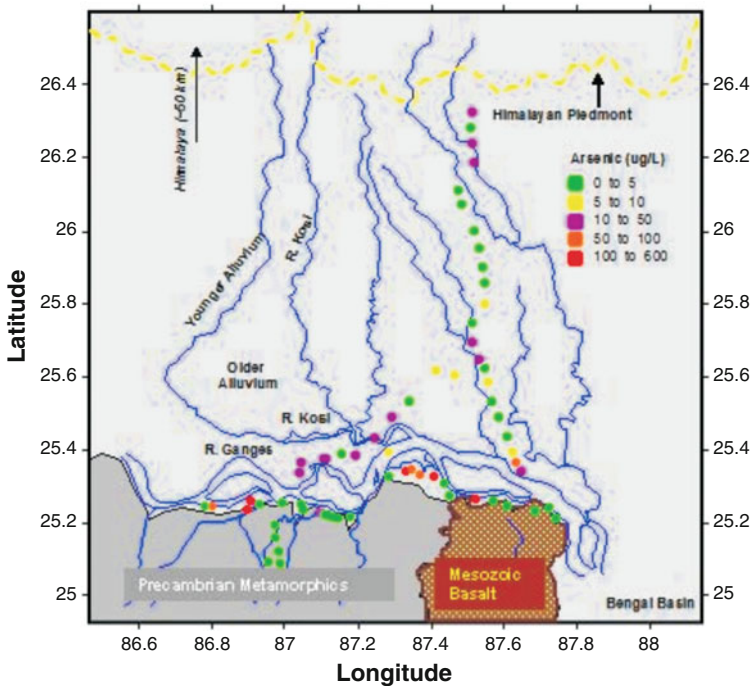
**Fig. 1.2** A conceptual cross-section along the line AA' shown in Fig. 1.1b, showing the probable disposition of the aquifers within the study area

cycles of fining upward sequence. Seismic studies have indicated the thickness of Quaternary Alluvium in the range of 300–400 m. The beds have gentle dip towards south. Within a depth of 100 m, the beds steeply dip toward south in the northern part of this fan deposit, and the layers are mostly composed of coarse sand and pebble with localized thin layers of clay. From north to south laterally continuous aquifers are present within a depth of 80 m bgl (Fig. 1.2). These are made up of coarse sand and gravels. The water levels observed during pre-monsoon ranged from 2.5 to 7.8 m

bgl (mean 4.2 m bgl). During mid-monsoon the difference between maximum (2.6 m bgl) and minimum (0.9 m bgl) water levels were found to be least. Mean water level during August is near surface ( $\sim 1.5$  m bgl) indicating a large part of post-August rainfall may be considered as rejected recharge. Average water level rise from monsoon recharge is  $\sim 3$  m. Between August and November, i.e. monsoon period  $\sim 37\%$  of the recharged water has been calculated to be seeping out as base-flow to rivers.

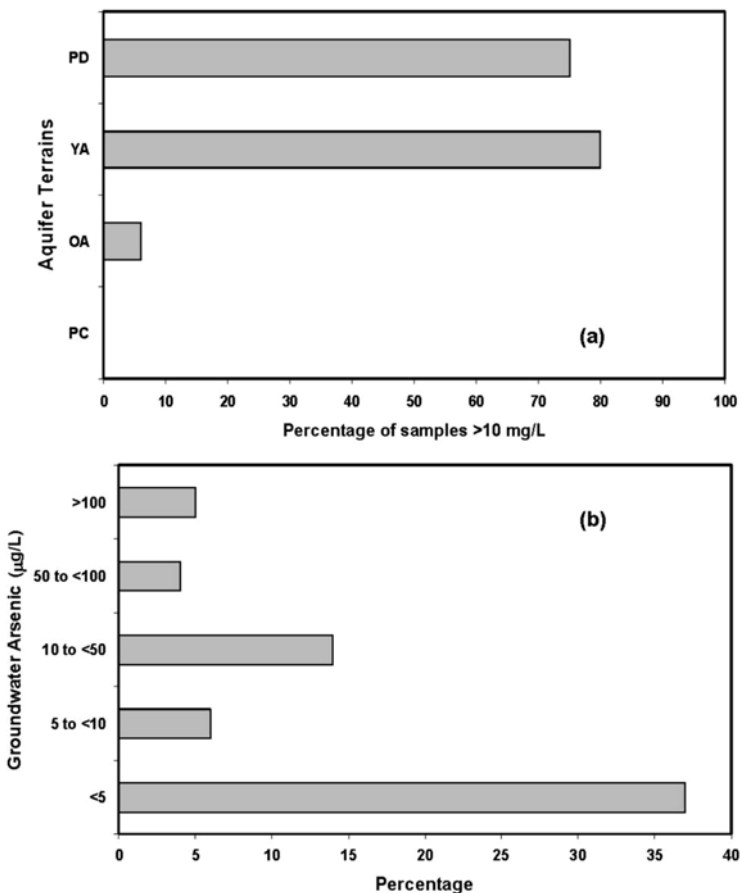
### 1.3 Methodology

Groundwater samples were collected from two transects (Fig. 1.3): (1) A 135 km long north-south transect extending from southern bank of the river Ganges up to the Indo-Nepal border in the north, through the river Kosi fan (groundwater sample  $n=38$ ); (2) A 90 km long east-west transect, running parallel to, and along the southern bank of the river Ganges. The groundwater sample collection and field measurements were done following standard procedures (e.g. Woods 1981). The groundwater samples are collected from hand-pumped tube wells and public water supply wells. The total depth intervals of the samples vary between 8 and 137 m.



**Fig. 1.3** Map showing the groundwater samples collected from the study area, classed by the arsenic concentrations detected in those samples

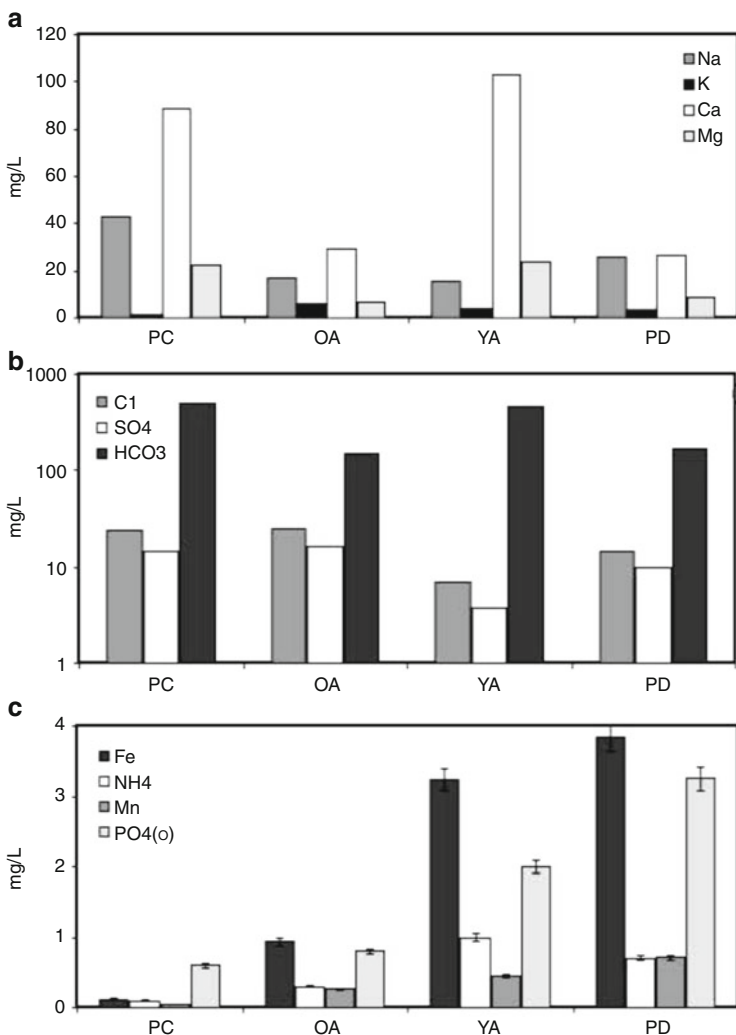
The groundwater sample locations in this study have been described following field information and other secondary information e.g. remote sensing. Accordingly, the samples have been classified into four geologic-geomorphic groups: aquifers composed of Pre-Cenozoic aged geologic media (comprising Precambrian metasediments and Rajmahal basalts,  $n=20$  [PC]), Alluvial sediments in the valleys of the river Ganges and its tributaries including Older Alluviums (the older flood plains and interfluvial plains,  $n=17$  [OA]) and Younger Alluvium (comprise the channel sediments and active flood plain deposits of rivers Ganges, river and other tributary systems,  $n=25$  [YA]), and Piedmont aquifers at the foothills of the Himalayas, irrespective of age or deposition agents,  $n=4$  [PD] (Figs. 1.3 and 1.4). Hence, the PD aquifer group does comprise characteristics of both younger and older alluvium of the numerous Himalayan first and second order tributaries of the river Kosi system.



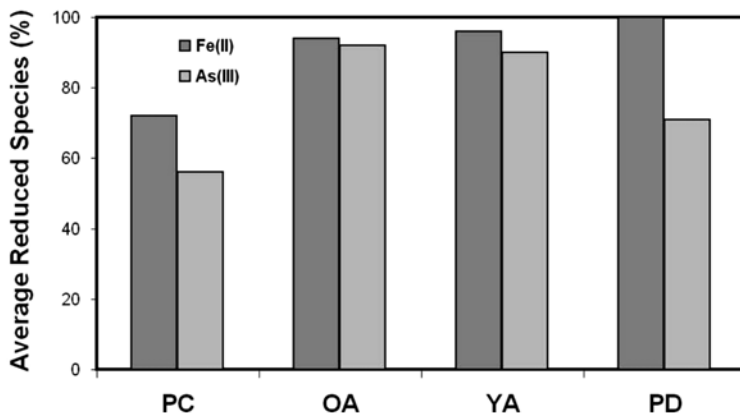
**Fig. 1.4** Plots showing the (a) percentage of the samples having groundwater arsenic concentrations  $\geq 10 \mu\text{g/L}$  in each of the aquifer terrains, and (b) percentage of samples within each concentration classes

### 1.4 Groundwater Chemistry

Here, we evaluate the different sources and mechanisms, which have led to the present composition of shallow groundwater for each of the terrains of the Central Gangetic Plain. The cations and anions in the groundwaters in the study area are probably sourced from silicate weathering, carbonate dissolution, and/or reactions like cation exchange (Figs. 1.5 and 1.6). Because of the known tropical, humid condition of the study area, presence of evaporite minerals and its dissolution seems to be impractical.



**Fig. 1.5** Plots showing median concentrations of (a) selected major cations, (b) selected major anions, and (c) selected minor ions present in the different aquifer terrains



**Fig. 1.6** Plot showing the percentage of the Fe(II) and As(III) redox species present within the total Fe and As concentrations detected in the groundwater samples of the different terrains

Groundwaters in all of the aquifers are mostly of Ca–HCO<sub>3</sub> facies with variants ranging from Ca–Na–HCO<sub>3</sub>–Cl to Na–HCO<sub>3</sub> types, suggesting evolution from water–rock interaction. To ascertain the sources of the major solutes Ca+Mg vs. total cations, were plotted to show distinct trends between the alluvial and non-alluvial aquifers (PC terrain). However, although the ratio of Ca+Mg to total cations tends to be higher for the YA group (medians of ratios ~0.7) than the other type of aquifers in the study area (the ratios are 0.34 for PD, 0.45 for OA and 0.57 for PC), the ratio for Na+K to total cations tends to be lowest for YA (median ratio=0.17 for YA), and median ratio ranges from 0.25 to 0.4 for other three groups. If the bivalent cations (e.g. Ca and Mg) in the YA groundwaters are derived from the silicate weathering, the provenance, i.e. the Himalayan source rocks would have been enriched in alkaline earth silicates e.g. Ca-feldspars, which are in contrary more enriched in orthoclase silicates, providing the flux of monovalent Na and K to the groundwater systems by silicate dissolution (Sarin et al. 1989; Galy and France-Lanord 1999; Mukherjee et al. 2009b).

The concentration of the total anion present in the PC and YA aquifer groundwater samples (>8,000 μM) are much more than of the total anion concentrations of the other groups, although in all of the aquifers, HCO<sub>3</sub><sup>-</sup> predominates the total anion budget. However, the concentrations of the anion in PC and YA (median >7,000 μM) are more than three times that of the OA and PD samples (median <3,000 μM). The median values for the molar ratio of total anion concentrations to HCO<sub>3</sub><sup>-</sup> in OA samples (<0.8) are less than that of YA samples (>0.9) suggesting that carbon cycling from primary mineral and/or secondary organic sources are a more dominant process in the YA aquifers than other aquifer groups. The median ratio of PC and PD aquifers are between that of OA and YA aquifer types. The HCO<sub>3</sub><sup>-</sup> versus Ca+Mg and Na+K bivariate plots suggest that the carbonate weathering has contributed to the HCO<sub>3</sub><sup>-</sup> in the YA and silicate dissolution contributing that to the PC aquifer groundwater. Generally, in the PC and OA aquifers, the Cl and SO<sub>4</sub><sup>2-</sup> concentrations are quite enriched than the YA and PD groundwaters.

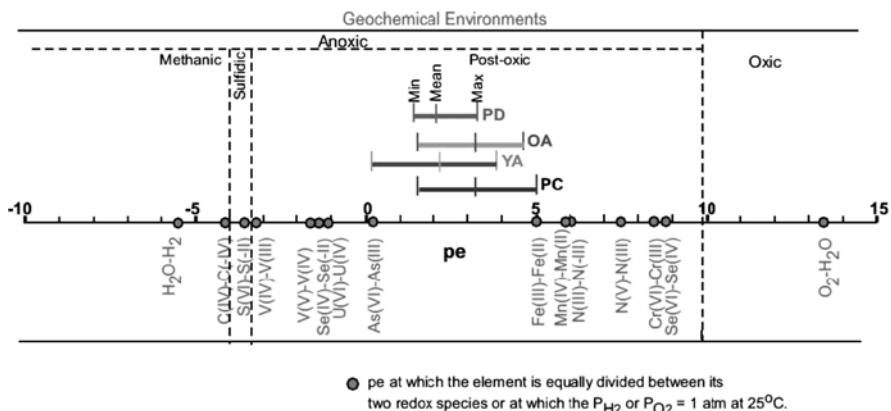
Bivalent cations ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) may have mobilized sorbed monovalent cations like  $\text{Na}^+$  and  $\text{K}^+$  from the aquifer matrix by cation exchange reactions for long residing groundwater in the study area, and the influence of the process can be estimated by subtracting equivalent concentrations of co-introduced anions ( $\text{HCO}_3^-$  and  $\text{SO}_4^{2-}$ ) from other processes of solute introduction like water-rock reactions. The  $\text{Na}^+$  leaching from the aquifer sediments are the residual from the subtraction of the equivalent concentrations of meteoric originated Cl from the Na (McLean and Jankowski 2000). A negative slope in bivariate plot (i.e.  $y = -x$ ) would suggest a possibility of active ion exchange reactions in the study area, as being observed in the case of PC samples, that have a slope of about  $-0.8$ . The proximity to the  $y = -x$  line might indicate that the long residing groundwater in the PC aquifers probably would have been exposed extensively to cation exchanges. On the contrary, the groundwater in the alluvial or piedmont aquifers can be conceptualized to have shorter groundwater residence time, along with dominance of shallow, local scale flow path in the shallow aquifers in higher gradient terrains of the study area, thus possibly translated to low to very low negative slope value, as indicator of lack of cation exchanges.

## 1.5 Groundwater Arsenic Distribution and Fate

The regional signature of the extent of groundwater As in the study area is not as pervasive as observed in the downstream fluvio-deltaic plains of the Bengal basin, and only  $\sim 35\%$  of the groundwater samples collected from the study area, irrespective of the aquifers, were detected to have dissolved As concentrations  $\geq 10 \mu\text{g/L}$  (WHO standard for safe drinking water), and only  $14\%$  were found to have As  $\geq 0.05 \text{ mg/L}$ . Groundwater As detection ranges from  $<1$  to  $520 \mu\text{g/L}$  (mean  $30 \mu\text{g/L}$  and median  $<5 \mu\text{g/L}$ ), if all the aquifers of the study area are taken together (Fig. 1.4). However, when classified by the separate terrains observed in the study from where the samples have been collected, even  $75\%$  samples for the recent alluviums and foothill deposits were detected to have As  $\geq 10 \mu\text{g/L}$ . In contrast there is none to negligible As detected in the groundwater collected from the aquifers of the PC and OA terrains. The YA aquifers are found to be most enriched in groundwater arsenic with an arithmetic mean of  $73 \mu\text{g/L}$  and median  $27 \mu\text{g/L}$ .

It is expected that because of the proximity, and similarity in lithology and groundwater systems, the As fate in the present study area existing in the upstream in the Ganges alluvial system should have some similarity with the downstream Bengal basin in terms of chemical characteristics and the spatial distribution of the As, along with its fate and transport. In order to understand the As fate, multivariate relationship have been evaluated in the study area, non-parametric Spearman correlations were calculated for As and other potentially influencing groundwater parameters of the samples suggested some interesting insight. These calculations indicate that different processes control the As fate in different terrains. For aquifers of all terrain, redox dependent mobilization of As is demonstrated by strong negative





**Fig. 1.7** Range of  $p_e$  values for the groundwater samples collected from each of the terrains along with their relative position in the geochemical environment (Modified after Mukherjee et al. 2012)

correlation with Eh ( $\rho = -0.44$  to  $-0.76$ ), as observable in reductive (Fe-Mn)OOH dissolution (Fig. 1.7). Such reduction dependent mobilization is also reinforced by correlation of As with reduction indicators like DO,  $SO_4$  and  $NH_4^+$  (Acharyya et al. 1999). It is expected that reductive dissolution of FeOOH onto which As is adsorbed, would lead to moderate to strong correlation between As and Fe. Such relationships have been advocated by workers like Nickson et al. (1998), Dowling et al. (2003), McArthur et al. (2004) and Stüben et al. (2003). However, in many other studies, such good correlation have not been observed (e.g. Swartz et al. 2004; Mukherjee and Fryar 2008; Mukherjee et al. 2008), probably suggesting the heterogeneity in the inter-dependence of the processes that might be controlling the co-existence of these elements. In the samples of this study, Fe correlates moderately to strongly with As with  $\rho = 0.38$  for YA and 0.66 for OA. However, for PD, Fe and As have very weak negative correlation ( $\rho = -0.12$ ). Similarly, while Mn and As show moderate to strong correlation for YA and OA ( $\rho = 0.32-0.47$ ), PD shows a negative correlation ( $\rho = -0.57$ ). In the contrary, competitive anions to As like  $HCO_3^-$  and ortho- $PO_4$  show very strong (for PD,  $\rho > 0.8$ ) or strong positive relationship (for OA,  $\rho = 0.53$  and 0.49, respectively), while they are moderate to insignificant for YA ( $\rho = 0.32$  and 0.14, respectively).

Such differences in correlations between the OA, YA and PD sites suggest that As is mobilized by multiple processes, in-situ or along the flow path. The YA and PD also have some correlation for As with pH ( $\rho = 0.48$  and 0.78), indicating probability of pH-dependent sorption reactions playing an influential role in regulating As mobility in the study area, which has not been suggested to have significant role in the Bengal basin (Mukherjee and Fryar 2008; Mukherjee et al. 2011). In summary it might be stated that the enrichment processes of dissolved As in the YA and PD might be influenced by considerably different mechanisms. While in YA, which are located mostly at the discharge ends of the flow paths, mobilization by metal-reductive dissolution is a potential dominant mechanism, enrichment in the recharge

areas, i.e. PD might be more influenced by competitive anion exchange between adsorbed As and ion introduced from mineral dissolution processes or competitive ionic exchanges by agricultural processes (e.g. ortho- $\text{PO}_4$ ).

## 1.6 Conclusion and Synthesis

It can be concluded from the present study and in earlier work (Mukherjee et al. 2012) that the water chemistry and elevated As distribution in the previously not-much-studied Central Gangetic plain and Kosi fan aquifers have some similarity, but more dissimilarity to that of the downstream, extensively studied, Bengal basin. The present study area is located in the eastern parts of the India subcontinent, stretching from the northern edge of the cratonic India to Himalayan foothills. Tectonically, thus the aquifers become part of the Himalayan foreland basin, mostly of stable shelf composition. The aquifers considered in the present study may be considered to be one of the best yielding aquifers of the subcontinent, and probably among some of the most prolific aquifers across the globe. Hence, the aquifers are most extensively agriculturally exploited. However, the concern about the reported As enriched groundwater from geogenic and non-point source has raised serious alarm in the huge population dependent on groundwater for their day to day needs. Moreover, the proximity of the immediately downstream, highly groundwater As enriched Bengal basin pose an intriguing question about the connection of the source and sedimentary history of the two adjoining basins. In this study, the objective was to delineate the As-enriched/prone aquifers of the Central Gangetic basin (including the Central Gangetic plain and Kosi fan). The hydrogeological techniques used in this study have shown that the groundwater in the aquifers within different host rocks in this basin suggests hydrochemical compositions that they have inherited from their geological and geomorphical evolutionary history. The aquifers in the study area can be generalized to be composed of the geologic material of Pre-Cenozoic aged rocks that comprise Precambrian metasediments and Cretaceous flood basalts, alluvial sediments in the valleys of the River Ganges and its tributaries including older alluviums in the older flood plains and interfluvial plains, and younger alluviums that are comprised of the channel sediments and active flood plain deposits of rivers Ganges, Kosi and other tributary systems, and Piedmont aquifers at the foothills of the Himalayas.

In general, the groundwaters existing in all of the aquifers are mostly of Ca-HCO<sub>3</sub> hydrochemical facies, with compositions ranging from Ca-Na-HCO<sub>3</sub>-Cl to Na-HCO<sub>3</sub> types. Like groundwater in many of the modern alluvial aquifers with wide water-rock interaction and organic load, HCO<sub>3</sub> is found to be the dominant anion. However, concentrations of anion in the PC and YA group are more than double that of the OA and PD groups, probably for totally different type of geochemical reactions. Much of the YA solutes in the groundwater are introduced from carbonate weathering reactions. However, the PD groundwater and many of the PC and OA samples are largely affected by silicate weathering by incongruent leaching

of the argillaceous metamorphics and volcanics in the Himalayas and the Pre-Cenozoic deposits, which act as a major solute provenance for groundwaters residing in these aquifers. Further, the potentially long residence time of groundwater in the PC terrain has resulted in to suspected cation exchange reactions, whereby monovalent cations got introduced from the host aquifer matrix. The PD samples are found to be more similar to OA in terms of major solutes composition but are more similar to YA groundwater in term of minor solutes.

The presence of redox sensitive species like As(III), Fe(II),  $\text{NH}_{3(\text{dis})}$ , and elevated  $\text{HS}^-$  indicates the occurrence of a reductive, post-oxic redox environment, with most groundwater samples being plotted in the metal reducing hydrogeochemical condition. Dissolved Fe concentrations in predominantly Fe(II) in groundwater for all terrains and As exist mostly as As(III). Reduction is more dominant in groundwaters in YA and PD aquifers, with signature of influence of agricultural recirculated water. Redox conditions (oxic to methanic, dominated by metal-reduction) are highly spatially variable, with no systematic depth-dependence. Almost 35 % of all of the collected groundwater from all of the four aquifers of the study area were cumulatively found to have dissolved As concentrations  $\geq 10 \mu\text{g/L}$ , of which, a major percentage (~45 %) have concentrations  $\geq 50 \mu\text{g/L}$ . However, when classified by the aquifer terrains, while the PC and OA groundwaters have none or negligible As, almost 75 % or more YA and PD groundwaters are enriched by As  $\geq 10 \mu\text{g/L}$ . The YA groundwaters are most enriched with As, with detected concentrations up to  $520 \mu\text{g/L}$ . Analyses of correlation between As and other redox parameters suggest that most of the As liberation and mobilization in the YA and OA aquifers are influenced by redox related mobilization, with suggested reductive dissolution of metal (Fe-Mn)-oxyhydroxides leading to reasonably good correlation of As with Fe. Interestingly, the piedmont groundwaters strongly correlate competitive anions that are probably influenced from anthropogenic activities like nutrients introduced by agricultural processes. Hence, the As fate in the regional groundwater recharge areas near the Himalayan foothills are influenced by mechanistically different geochemical methods than the groundwater residing in the discharge areas typified by extensive alluvial deposits in vicinity of the modern river channels. Hence, in the present study area of Central Gangetic Basin that is located upstream of the Bengal basin or the Lower Gangetic plain and delta, hydrochemistry are much more variable in the solute distribution and fate as a function of differently evolved geological and geomorphic terrains. In comparison, the Bengal basin aquifers have overlapping facies aquifer systems with obscure terrains divisions. Consequent to the huge active sedimentation in that basin, most of the aquifers are geochemically similar to composition of the younger alluviums, with similar distribution and fate of As like the YA aquifers of the present study area.

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# Chapter 2

## Assessment of Subsurface Lithology by Resistivity Survey Coupled with Hydrochemical Study to Identify Arsenic Distribution Pattern in Central Gangetic Plain: A Case Study of Bhagalpur District, Bihar, India

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### 2.1 Introduction

The occurrence of arsenic (As)-rich alluvial groundwater is a worldwide problem (Kumar et al. 2010). Most studies of As pollution have focused on the predominance of As poisoning in the groundwater of West Bengal (India) and Bangladesh and thought to be limited to the Ganges delta i.e. the lower Gangetic plain (Bhattacharya et al. 1997; Ahmed et al. 2004; Ben et al. 2003). Some states as Uttar Pradesh and Bihar reported the presence of elevated concentrations of arsenic in drinking water wells sporadically (Acharyya and Shah 2004; Chakraborti et al. 2004; Acharyya 2005; Chauhan et al. 2009; Sankaramakrishnan et al. 2008; Srivastava et al. 2008; Kumar et al. 2010). Several authors suggested that the reductive dissolution of Fe (III)-oxyhydroxides in strongly reducing conditions in the young alluvial Holocene sediments is the cause for arsenic mobilization (Harvey et al. 2002; Nickson et al. 1998; Nickson et al. 2000). Groundwater quality is controlled by various factors viz. composition of recharging water, the mineralogy and

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