CHAPTER 1

Introduction

1.1. General background

Arsenic (As) and fluoride (F⁻) are two of the most toxic inorganic contaminants which can occur in natural water. Epidemiological studies have shown that inorganic As is a serious toxicant and can cause a variety of adverse health effects, such as dermal changes, respiratory, pulmonary, cardiovascular, gastrointestinal, haematological, hepatic, renal, neurological, developmental, reproductive, immunologic changes leading to cancer and other degenerative effects of the circulatory and nervous system [1, 2, 3, 4]). In view of the above factor World Health Organization in 1993 lowered the earlier permissible limit of As in drinking water from 50 μ gL⁻¹ to 10 μ gL⁻¹ [5]. Cases of endemic arsenicosis or As poisoning have been reported from countries like Argentina [6, 7], Bangladesh [8-10], Cambodia [11-13], Vietnam [11, 14], Pakistan [15, 16] and India [17-20].

Fluoride on the other hand has been found to have a debilitating effect on human physiology, leading to both dental and skeletal fluorosis which cripples the body and diminishes a person's ability to continue daily activities. The WHO has prescribed 1.5 gL⁻¹ as the maximum daily intake limit for F⁻ in drinking water. Endemic fluorosis has been reported to be a problem in many parts of the world like Africa [21-25], India [25, 26], China [25, 27], Japan [25, 28], Canada [25, 29], Turkey [25, 30] and even developed countries like the USA [25, 31].

When the threats are anthropogenic there is a chance or a choice to reduce or mitigate the problem; however when nature itself is responsible for such a calamity, it becomes very difficult to deal with the problem. Both As and F^- have been found to be primarily geogenic in origin, and the two contaminants have been recorded in a number of different environments. However in India the two contaminants have been found to occur in different niche environments. Fluoride was detected in the groundwaters of the

arid and semi arid region like Andhra Pradesh, Tamil Nadu, Rajasthan, Orissa, Karnataka, Bihar, Punjab, Uttar Pradesh and Gujarat [32-36]; while high groundwater As has been detected in areas with fluvial environments where precipitation and groundwater recharge rates were found to be high. The table 1.1 depicts the occurrence of As in some of the states of India [37]. It can be observed that occurrence of high groundwater As is limited mainly to the states where fluvial environment has been detected. The Brahmaputra flood plains (BFP) lie in the state of Assam, while Middle Gangetic Plains (MGP) is spread over the states of Bihar and Uttar Pradesh and West Bengal lies in the lower Ganga plains. The state of West Bengal and the MGP have been reported to be the two most severely As affected regions of India which have alluvial settings [38- 40]. In recent years both the contaminants have also been reported to occur in the groundwater of the North-Eastern part of India [41, 42].

Although a lot of work has been done on different aspects of both the contaminants like their hydrogeology, biochemistry etc., yet studies on their co-contamination perspective are lacking. Very few works around the world have actually delved into this matter. Actual laboratory simulations like sorption and desorption experiments and their comparison with the natural occurrence of As and F⁻ are reported to be rare. In India groundwater As and F⁻ have been studied extensively but studies on their cocontamination behaviours are lacking. In the North Eastern part of India, very few people have actually studied the hydrogeochemical aspect of the two contaminants even on individual levels, and co-contamination studies are completely absent. Characterization and understanding of the different phases or pools of As in the sediment and soils is entirely lacking. The problem of As and F^{-} co-contamination is important because both have been found to have a worldwide distribution and their potential health hazards are severe, moreover the probable interactions that the two might exhibit in the human physiology are not totally understood. Therefore the need of the hour is to focus on the co-behaviour of both the contaminants to get a better insight into their interactive behaviour.

STATE	No of wells	As	Fe	Salinity	NO ₃
	analyzed			-	
Andhra Pradesh	3875	0.00	3.54	25.68	8.67
Assam	14645	3.86	96.06	0.00	0.00
Bihar	14068	7.76	77.82	0.00	0.01
Chhattisgarh	7191	0.00	92.45	2.78	0.00
Gujrat	366	0.00	0.00	0.27	53.28
Haryana	15	0	0	0	0
Jharkhand	100	0	79	0	0
Karnataka	5123	0.43	16.49	10.68	24.50
Kerela	928	0.00	62.82	18.10	7.22
Madya Pradesh	2888	0.00	6.82	2.74	0.00
Maharastra	1659	0.00	19.11	17.18	32.31
Orissa	10264	0.00	87.97	7.81	0.11
Punjab	262	0.00	87.40	2.67	0.00
Rajesthan	24437	0.00	0.05	64.91	5.85
Tamil Nadu	670	0.00	83.13	11.94	2.69
Uttar Pradesh	946	16.38	8.14	42.39	0.11
Uttarakhand	49	0.00	85.71	0.00	8.16
West Bengal	3190	18.93	77.46	0.09	0.00

 Table 1.1: State- wise representation in percentage of total groundwater wells

 analyzed that were affected with arsenic contamination

Source: Ministry of drinking water and sanitation, 2014.

* Database for Delhi is not available but both As and F⁻ contamination are reported in this region.

1.2. Problem statement

The Brahmaputra flood plains (BFP) of Assam have been described as the lifeline for the people of Assam and also the rest of North East India. Most of the agricultural and industrial activities are concentrated in this region, and almost all the important cities and towns of Assam including Guwahati, the largest city of the state are situated in the BFP region. Thus the BFP is very densely populated. The North Eastern states including Assam have been found to be the latest additions in India with high groundwater As and F⁻ [42]. The As and F⁻ contamination has been reported to be very extensive in the state of Assam, where 21 of the 27 districts have reported high As, while high groundwater F⁻ has been found in drier rain shadow regions like Karbi Anglong and Nagaon districts [42]. Groundwater abstraction has been found to be very high in the BFP region of Assam, which is used mainly for drinking, industrial and agricultural practices. Moreover the usage of fertilizers and other agricultural practices have reportedly increased over the years, which could impact the mobilization process of As and F⁻ in the BFP. Sufficient knowledge on the hydrogeochemistry of both the contaminants is lacking in the region, yet it can be said that As and F^- contamination of groundwater in the BFP is bound to increase in the future in part due to the change in land use patterns of the region.

1.3. Research question

The main questions that this research work attempts to answer are i) how As and F⁻ can co-exist and interact with each other in a natural setting as well as in controlled conditions, ii) how As and F⁻ are released into aqueous solution from solid and adsorbed species and iii) what factors control such release and if the two are released from the solid phase, what will be the fate of their aqueous species in presence of each other. The behaviour of As and F⁻ is controlled by a number of factors. Both exist as anions, therefore the very idea of a study is an interesting prospect to see whether they behave synergistically or antagonistically in the system (Fig. 1.1) and what relation they could exhibit with other parameters.



Figure 1.1: Schematic diagram of probable As-F⁻ interaction

Another important research question that we wanted to address was whether or not proximity to the river had any impact on the distribution and occurrence of As and F^- , because the two contaminants have been found to occur in regions with different

environmental and hydrogeological conditions. It has already been mentioned in the "background" section that in India high groundwater As has been found in fluvial regions with high sedimentation and aquifer recharge rates. Fluoride on the other hand has been found to occur in drier regions with more rock-water interaction and slower groundwater recharge [25, 43- 45] found high As in the fluvial regions of Hanoi, Vietnam.



Figure 1.2: Map of the Hanoi area depicting arsenic, groundwater heads, peat and ammonium levels Berg et al [45] Effects of proximity on groundwater As level is clearly visible

Closer examination of the above study showed that all the groundwater samples with high As levels were found at close proximity to the Red River (Fig. 1.2). A similar observation can also be made from the study of Shah [46]; in this study groundwater

samples with higher As were located close to the Ganga River in the MGP (Fig. 1.3). In a study conducted on groundwater F^- by [47] in the state of West Bengal, India, very low levels of F^- were detected indicating that fluvial environment which is prevalent in West Bengal was not suitable for occurrence of high groundwater F^- . Different studies have shown that F^- has a positive relation with NaHCO₃⁻ type water [48, 49] while As has also been shown to rise under alkaline pH [50].



Figure 1.3: Map showing arsenic afflicted regions in parts of Uttar Pradesh [46]. Proximity plays important role in groundwater distribution

It is very important to fill the existing gaps especially those mentioned in the "background" section like absence of proper laboratory simulation to study the interactive behaviour of the two contaminants. Such a study is essential to answer questions like whether there will be any differences observed between the natural process of mobilization and laboratory based processes of sorption and desorption of the two contaminants. Last but not the least a very important question which this study

attempts to find an answer to is: Will anthropogenic activities impact the process of As and F^- contamination? Considering the aforementioned research questions, this study was titled "Understanding arsenic phase distribution and co-contamination perspective with fluoride in the Brahmaputra flood plains, Assam, India" in an effort to develop deeper understanding of the extent and the mechanism of As and F^- co-contamination in the BFP.

1.4. Aims and objective

Under the preview of the above discussion, the five field objectives of this study were to:

- I. Identify the degree of co-occurrence of As and F⁻ in the entire BFP and its temporal/seasonal fluctuations.
- II. Evaluate the effect of river proximity and enrichment processes for As and F⁻.
- III. Assess the As-F⁻ co-contamination perspectives in the natural and laboratory setup with an emphasis on identification of the governing factors.
- IV. Understand the phase distribution of As in the soil and sediments of the BFP and observe As and F⁻ co-contamination under laboratory settings.
- V. Integration of the results of water quality, phase distribution of As in solid system through sequential extraction and batch dissolution experiments.

1.5. Research design

Figure 1.4 represents the schematic diagram of our research design. In order to conduct a study with a fruitful outcome in a region as vast as the BFP, a preliminary literature survey on prevailing conditions like groundwater quality, minerals, hydrogeology and sediment types was considered very effective. An initial sampling of the groundwater and the sediments can be useful to observe the temporal and spatial distribution of the two contaminants and their hydrogeochemistry. However limited number of samples over a large area can be problematic as many local variations may not be observed. This problem was tackled by conducting a micro scale study on a smaller area (Jorhat district) of the much larger BFP region to observe missing trends. While sampling, proper placement of groundwater sampling locations can go a long way in ensuring observation of important trends. We have already discussed the aspect of groundwater As level with river proximity. To study proximity to the river has any effect on F⁻ distribution or not was considered as one of the foci of this study. Therefore the groundwater samples were strategically collected by taking into account the distance of the sampling points from the river.

In order to derive a suitable idea and propose a hypothesis we studied the behavioural patterns of As and F^- by observing their interactions with each other and with other physico-chemical parameters. However the empirical relations observed between As and F^- and their relations with other hydrogeochemical parameters like the major ions were not enough to give a proper idea for generating a hypothesis. In order to counter the above problem and arrive at a suitable conclusion we decided to observe the relation of both As and F^- with each other and with the different parameters of their source materials i.e., the soils and sediments, in a controlled laboratory environment. The distributions of the As phases or fractions in the source materials were observed before any batch desorption experiment was carried out to observe the relation between the As phases and the desorption process.



Figure 1.4: Schematic diagram for the design of research in our study

1.6. Thesis integration

Figure 1.5 gives the schematics and the flow of the chapters in this thesis. A brief summary has been added after the diagram for a better insight into each chapter.



Figure 1.5: Organization of the thesis

Chapter 1: The first chapter briefly describes this doctoral work where the background of the problem has been discussed. The problem of As and F^- co-contamination in context of the BFP has been introduced in this chapter. This chapter raises the research question and the motivation to conduct this research. It also discusses the aims and the objectives; and the research design. The chapter concludes with a brief description and novelty of this work.

Chapter 2: The second chapter gives a detailed account of the literature which was surveyed for conducting this research. This chapter discusses the findings and the reporting of previous works which were considered important and relevant during the course of our own study.

Chapter 3: This chapter discusses the relevant resources which were used during this study. The chapter gives a detailed account of the study area, and describes the methods used and the protocols followed for the technical and analytical aspect of this work.

Chapter 4: This chapter is a spatio-temporal study of As and F⁻ in the study area. The chapter is divided into three parts, the first deals with spatial distribution of As and F⁻ in the larger BFP region, the second focuses on the influence of river proximity on As and F⁻ distribution in the BFP and the third focuses on the macro and micro scale comparative study of As in the BFP.

Chapter 5: This chapter introduces the problem of As and F^- co-contamination, and discusses the relation of As and F^- with each other and with other physico-chemical parameters to assess the co-contamination perspective in the natural setting. The chapter uses an integrated approach of major ion chemistry, multivariate statistical techniques and speciation modelling to study the interaction between the two groundwater contaminants.

Chapter 6: The previous chapter leads to sixth chapter where we add the dimension of sequential extraction of As and batch sorption experiment. The former was used to identify the "operationally defined" fractions of As which could have notable implications in the behaviour of As and F^- in the BFP. Desorption experiment was conducted to observe the co-evolution of As and F^- in a laboratory setting.

Chapter 7: The final chapter summarises the work of all the chapters and presents the hypothesis for this work. It also discusses the scope of the work and its future implications.

1.6. Highlights of this work

I. Previous studies on As and F⁻ hydrogeochemistry are very limited and lacking in the BFP.

- II. The BFP has not been studied in its entirety in previous works dealing with groundwater As and F⁻, our work is a pilot attempt for doing the same.
- III. Co-contamination of As and F⁻ has not been studied in the BFP. The present work has attempted to do the same by comparing the observations in the natural settings and laboratory experimentation by sorption and desorption studies.
- IV. Soil and sedimentary characterization and its relation to As phase or fraction distribution is lacking in the BFP region. The present study has attempted to do the same and tried to relate it to the process of As and F⁻ co-contamination.

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