

**Department of Forest - and Soil Sciences**

Institute of Soil Research



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**ARSENIC CONTAMINATION OF SOILS AND  
AGRICULTURAL PLANTS THROUGH IRRIGATION WATER  
IN NEPAL**

**Dissertation**

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Submitted by  
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## Summary

This study was conducted in the irrigation area of Nawalparasi district which is known as highly contaminated by arsenic among the Terai districts of Nepal. The purpose of this study was to analyse the arsenic of agricultural plants due to irrigation with arsenic contaminated water and the role of phosphorus fertilization. It is hoped that the result of this study will help to develop guidelines to protect the food chain by arsenic contamination in Nepal which is an important issue of the Millennium Development Goal (MDG) regarding the food security.

65 % of all 52 wells showed an arsenic concentration higher than  $0.05 \text{ mg L}^{-1}$ , which is the Nepal interim standard for drinking water. Overall, 36 % of the irrigation wells exceeded the FAO guideline value ( $0.1 \text{ mg L}^{-1}$ ) for arsenic in irrigation water. A wide range of arsenic concentrations was observed in the irrigation water, ranging between  $< 0.05$  and  $1.014 \text{ mg L}^{-1}$ . The As(III) dominates in the water with a mean percentage of about 76%.

The arsenic concentrations in the irrigated soils ranged from  $6.1$ - $16.7 \text{ mg kg}^{-1}$ . The arsenic content was significantly ( $p < 0.05$ ) higher in the soils of the vegetable fields than in the soils of the rice fields. The arsenic content in different parts of plants decreased in the order of roots  $>$  shoots  $>$  leaves  $>$  edible parts. The mean arsenic content of edible parts (dry weight) decreased in the order of onion leaves ( $0.55 \text{ mg kg}^{-1}$ )  $>$  onion bulb ( $0.45 \text{ mg kg}^{-1}$ )  $>$  cauliflower ( $0.33 \text{ mg kg}^{-1}$ )  $>$  rice ( $0.18 \text{ mg kg}^{-1}$ )  $>$  brinjal ( $0.09 \text{ mg kg}^{-1}$ )  $>$  potato ( $< 0.01 \text{ mg kg}^{-1}$ ).

The arsenic contents of different soil aggregate fractions and its correlation with Fe, Mn, Al and Ca were studied in a rice and a vegetable field. The investigated aggregate size fraction was 1 – 2 mm. Arsenic content was highest in the smallest aggregates. Under rice no distinct correlation of arsenic with Fe, Mn, Al and Ca was found. Under vegetables As showed a good correlation with Fe, Mn and Ca in the smaller aggregate fractions ( $\leq 0.63 \text{ mm}$ ).

The laboratory experiment showed that the arsenic solubility was highly influenced by changes in the soil redox potential. The As solubility was increased about 5 fold after the addition of Di-Ammonium Phosphate (DAP) fertilizer under redox potential conditions (aerobic to anaerobic) while solubility of Fe and Ca was not affected. This indicates that P-fertilizer can solublize As through ion exchange processes.

## Zusammenfassung

Zielsetzung dieser Arbeit war die Untersuchung der Arsenbelastung von Nahrungspflanzen in Nepal durch arsenkontaminiertes Bewässerungswasser sowie von zusätzlichen Problemen durch die Anwendung von Phosphordüngemitteln. Sowohl die Feld- wie die Laboruntersuchungen zeigten, dass der Arsengehalt der Pflanzen wesentlich vom Arsengehalt des Bewässerungswassers abhängt, jedoch auch von der Art der Pflanzen und der Arsenkonzentration in der Bodenlösung. Darüber hinaus zeigten spezielle Laborexperimente, dass der höchste Arsengehalt in den kleinsten Bodenaggregaten vorkommt und dieser wiederum sehr stark von den Redox-Bedingungen des Bodens, von der Bewässerungstechnik (mit oder ohne Wassereinstau) abhängt. Während bei Wassereinstau in Reiskulturen keine klare Korrelation zwischen dem Gehalt an Arsen, Eisen, Mangan, Aluminium und Kalzium in den Bodenaggregaten gefunden werden konnte, zeigte der Arsengehalt in Böden mit Gemüsebau (ohne Wassereinstau) deutlich, dass Arsen in den kleinen Bodenaggregaten ( $\leq 0,63$  mm) Komplexe mit Eisen, Mangan und Kalzium bildet. Die Ausbringung von Di-Ammoniumphosphat-Dünger erhöhte die Arsenlöslichkeit sowohl unter aeroben wie anaeroben Bodenbedingungen um das fünffache.

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# Arsenic contamination of soils and agricultural plants through irrigation water in Nepal

## Chapter 1

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### 1 Introduction

#### 1.1 Arsenic

Arsenic is a naturally occurring element in bedrock and soil (Gerba, 1996). It is a metalloid and usually found in the environment combined with other elements, such as oxygen, chlorine and sulfur, or in organic form. Inorganic arsenic occurs naturally in many kinds of rock, especially ores that contain copper, lead, iron, nickel, and other metals. The average level of arsenic is  $1.8 \text{ mg kg}^{-1}$  in the earth's crust, normal background concentration is  $0.2\text{-}15 \text{ mg kg}^{-1}$  in the lithosphere, less than  $15 \text{ mg kg}^{-1}$  in soils with a mean value of  $5 \text{ mg kg}^{-1}$  (Bowen, 1979),  $0.02\text{-}2.8 \text{ ng m}^{-3}$  in the atmosphere, and less than  $1 \text{ }\mu\text{g L}^{-1}$  in aquatic environment. Arsenic can exist in four valency states:  $-3$ ,  $0$ ,  $+3$  and  $+5$ . Under reducing conditions, arsenite [As(III)] is the dominant form; arsenate [As(V)] is generally the stable form in oxygenated environments. Elemental arsenic is not soluble in water but its salts exhibit a wide range of solubility depending on pH and the ionic environment.

Arsenic is present in more than 200 mineral species, the most common of which is arsenopyrite. It has been estimated that about one-third of the atmospheric flux of arsenic is of natural origin. Volcanic action is the most important natural source of arsenic, followed by low temperature volatilization. Inorganic arsenic of geological origin is found in groundwater used as drinking water in several parts of the world, for example Bangladesh, India and Nepal. Organic arsenic compounds such as arsenobetaine, arsenocholine, tetramethylarsonium salts, arsenosugars and arsenic-containing lipids are mainly found in marine organisms although some of these compounds have also been found in terrestrial species.



### **1.1.1 Arsenic in water**

The concentration of arsenic in natural waters, including groundwater, is usually below  $10 \mu\text{g L}^{-1}$ . However, arsenic mobilization is favored under some specific hydrogeochemical conditions, especially highly reducing (anaerobic) conditions, which can bring about the dissolution of iron oxides and the associated de-sorption of arsenic. One of the key hydrogeochemical advances of the last few years has been in the better understanding of the diverse mechanisms of arsenic mobilization in groundwater, as well as its derivation from different mineral sources. The most important mineral sources in aquifers are metal oxides (especially iron oxides) and sulfide minerals (especially pyrite). Release of arsenic from sediments to groundwater can be initiated as a result of the development of highly reducing (anaerobic) conditions, leading to de-sorption of arsenic from iron oxides with the breakdown of the oxides themselves. Such reducing conditions are usually found in recently deposited fine grained deltaic and alluvial sediments. Release of arsenic can also occur in acidic groundwaters under oxidizing (aerobic) conditions. This tends to occur in arid and semiarid settings resulting from extensive mineral reaction and evaporation.

Due to the carcinogenic nature of arsenic, the World Health Organization (WHO) recommends a provisional guideline value for arsenic in drinking water of  $10 \mu\text{g L}^{-1}$  (WHO, 2001), which has been adopted by most industrial countries. Most developing countries still use the former WHO recommended maximum permissible limit for arsenic of  $50 \mu\text{g L}^{-1}$  (WHO, 1984) as their national standard, due to economic considerations and the lack of tools and techniques to measure accurately at lower concentrations. Groundwaters of 20 countries were found to be contaminated with arsenic (Rahman et al., 2006). Mostly the countries of south Asia are highly affected by the arsenic contamination in ground water.

### **1.1.2 Arsenic in soil**

Most natural soils contain low levels of arsenic, but industrial wastes and pesticide applications may increase concentrations. Naturally elevated levels of arsenic in soils may be associated with geological substrata such as sulfide and iron ores mainly realgar

(AsS), orpiment ( $\text{As}_2\text{S}_3$ ), arsenopyrite ( $\text{FeAsS}$ ), and lollingite ( $\text{FeAs}_2$ ). But contamination of soils by anthropogenic activities can have concentrations of arsenic up to several grams (Van Zwieten et al., 2003; NAS, 1977). Arsenic concentrations of up to 23 800  $\text{mg kg}^{-1}$  were reported in soils contaminated with mine or smelter wastes (Alvarez et al., 2006). Chatterjee & Mukherjee (1999) reported arsenic levels of 20 100 – 35 500  $\text{mg kg}^{-1}$  in soil around the effluent dumping point of an arsenical pesticide manufacturing plant in south Calcutta. Soil on agricultural land treated with arsenical pesticides may retain substantial amounts of arsenic. Mean total arsenic concentrations of 50–60  $\text{mg kg}^{-1}$  have been recorded for agricultural soils treated with arsenical pesticides (Takamatsu et al., 1982). Further more the use of arsenic contaminated water in irrigation also is a potential source of arsenic in agriculture soil.

Arsenic is emitted into the atmosphere by high-temperature processes such as coal-fired power generation plants, burning vegetation and volcanism. Natural low-temperature biomethylation and reduction to arsines also releases arsenic into the atmosphere. Arsenic is released into the atmosphere primarily as  $\text{As}_2\text{O}_3$  and exists mainly adsorbed on particulate matter. These particles are dispersed by the wind and are returned to the earth by wet or dry deposition. Arsines released from microbial sources in soils or sediments undergo oxidation in the air, reconvertng the arsenic to non-volatile forms, which settle back to the ground. In well-oxygenated water and sediments, nearly all arsenic is present in the thermodynamically more stable pentavalent state (arsenate). Some arsenite and arsenate species can interchange oxidation state depending on redox potential (Eh), pH and biological processes. Some arsenic species have an affinity for clay mineral surfaces and organic matter and this can affect their environmental behaviour. There is potential for arsenic release when there is fluctuation in Eh, pH, soluble arsenic concentration and sediment organic content. Weathered rock and soil may be transported by wind or water erosion. Many arsenic compounds tend to adsorb to soils, and leaching usually results in transportation over only short distances in soil.

### 1.1.3 Arsenic in Plants

The arsenic is readily taken up by plants, the degree of uptake being dependent on soil pH (Gerba, 1996). The arsenic content of plants grown on soils that had never been treated with arsenic-containing pesticides varied from 0.02 to about 5 mg kg<sup>-1</sup> DW. Plants grown on arsenic contaminated soils may, however, contain considerably higher levels, especially in the roots. Plants growing on arsenical mine wastes (south-west England, UK) contained mean arsenic levels ranging from 350 to 2 040 mg kg<sup>-1</sup> DW; a maximum concentration of 6 640 mg kg<sup>-1</sup> was reported for *Jasione montana* (Porter & Peterson, 1975). Benson et al. (1981) reported mean arsenic concentrations of 1 480 and 1 070 mg kg<sup>-1</sup> DW for the grasses *Agrostis stolonifera* and *Agrostis tenuis* growing on arsenical mine waste. Jonnalagadda & Nenzou (1997) reported arsenic concentrations in couch grass (*Cynodon dactylon*) growing on or near gold-arsenic mine dumps (Zimbabwe) ranging from 200 to 1 660 mg kg<sup>-1</sup> DW in stems and from 1 020 to 10 880 mg kg<sup>-1</sup> in roots. Mean concentrations of arsenic in the leaves of plants growing near a copper mine (northern Peru) ranged from 111 to 1 651 mg kg<sup>-1</sup> DW (Bech et al., 1997). The arsenic content in agricultural plants grown on arsenic contaminated soil has reported throughout the world (Duxbury et al., 2003; Huang et al., 2006; Sapunar-Postruznik et al., 1996). Similarly, the arsenic enters into the food chain due to the irrigation with arsenic contaminated water. A study done in Bangladesh showed that some vegetables contained higher amounts than the recommended limit of 1 mg kg<sup>-1</sup> set in Australia (Das et al., 2004).

## 1.2 Arsenic situation in Nepal

The studies on arsenic in drinking water in Nepal started in 1999 by the Department of Water Supply and Sewerage (DWSS) with financial support of the World Health Organization (WHO). Consequently, this study showed that Nepal has a problem of arsenic in drinking water. Several government and non-government organizations, which are working as water suppliers, have been involved in testing arsenic in drinking water and to combat this problem. Till the date about 737 009 tubewells, used to extract drinking

water, have been tested for arsenic out of 1 000 000 (NASC, 2007; Personal communication). The results show that 2.3% of tubewells exceeded  $50 \mu\text{g L}^{-1}$  (Nepal Interim Standard for arsenic) and 10.3% tubewells contained arsenic above  $10 \mu\text{g L}^{-1}$  (WHO guideline value). Among 20 Terai districts, Nawalparasi district was reported as a highly arsenic affected area where 12.3% of the tubewells contained arsenic above  $50 \mu\text{g L}^{-1}$  and 26% contained above  $10 \mu\text{g L}^{-1}$ .

About 3 million hectares (approx. 20% of total land of the country) is farmland (Priyar, 2005; MOFSC, 2002; Sainju & Thapa, 2000) in Nepal. Out of the 3 million hectares, 46.6% is in Terai (Panta & Belbase, 2003). The Terai, is a flat extension of the southern Indo-Gangetic plain with altitudes ranging from 66 m to 300 m. Although there is a high potential of availability of surface water for irrigation, it could not be used due to a lack of infrastructure. Only half of the farmland of the Terai has irrigation facilities. Therefore, the use of groundwater is increasing day by day because the government of Nepal has given high priority to groundwater irrigation schemes through the provision of subsidies.

Concerning the water used for irrigation, Cemat Water Lab-Nepal was collected and tested the water samples from tubewells which are specially installed to extract groundwater for irrigation with support of the World Bank /Nepal in May 2003. Under this program 522 tubewells were monitored, among which 485 (93%) have arsenic less than  $10 \mu\text{g L}^{-1}$ , 32 (6%) tubewells have  $11-50 \mu\text{g L}^{-1}$  and 5 (1%) tubewells have an arsenic content higher than  $50 \mu\text{g L}^{-1}$  (Chaturvedi, 2003). Even considering that this study could not cover all tubewells installed for irrigation, it revealed that the irrigation water has been contaminated by arsenic. Therefore, many farmers in Nepal are irrigating with arsenic containing water for growing crops and vegetables. But in Nepal, until today there are no data available regarding the arsenic content in edible parts of crops and vegetables grown with arsenic contaminated irrigation water, affecting the health of people due to their consumption.

## Chapter 2

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## 2 Objective and reasons of the research topic

### 2.1 Objectives

The general goal of this study is to know the arsenic content of edible parts of crops/vegetables grown with arsenic contaminated irrigation water.

#### Specific Objectives

- To screen the arsenic content in irrigation water;
- To generate data for arsenic contents in cultivated soil;
- To compare the arsenic content in different parts of crops and vegetables (root, stem, leaf and grain) grown through irrigation water with highly and low arsenic contamination;
- To know the distribution of arsenic in soil aggregate fractions;
- To know the effect of the redox-potential of soils and the application of Di-Ammonium Phosphate (DAP) fertilizer on the arsenic sorption and de-sorption in soils.

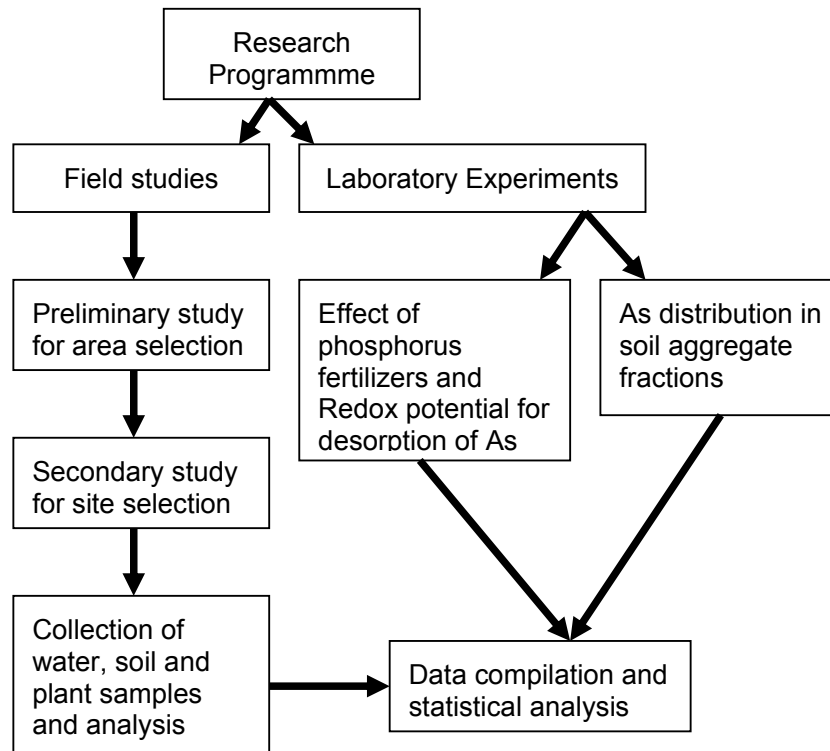
### 2.2 Reasons of the research topic

A census carried out in 2001 indicated that about 11 million people are living in the Terai of Nepal. Many arsenic contaminated tubewells are used for irrigation in this region. Therefore there is the danger of arsenic contamination of the food chain. This means that people are at risk of arsenic intoxication. This has raised great concern among the local people, the government as well as various non-governmental organizations involved in the health sector in Nepal. Except for the direct effect of arsenic contaminated drinking water on human health, the studies about arsenic phytotoxicity are completely lacking in Nepal. An investigation on this subject is, therefore, desperately needed in order to know the extent of arsenic content in crops/vegetables consumed by local people, which could be an important information for further research related to health effects of arsenic through the food chain.

### 3 Materials and Methods

#### 3.1 Research programme

The different steps of the research programme are shown in fig 3.1.



*Fig. 3.1. Schematic presentation of the research programme*

#### 3.2 Field studies

Three Village Development Committees (VDCs) of one district: Manari, Devgaoun and Sukrauli were selected for the study. Manari and Devgaoun are neighbouring VDCs and Sukrauli lies about 10 km west of them. About 150 seesaw wells were in use at the time of the study.

##### 3.2.1 Selection of wells and water sample collection

At first arsenic concentration in the water samples of all available seesaw wells which are used for irrigation purpose were measured by using ENPHO field test kits to identify

seesaw wells for research. Then 52 seesaw wells were selected for further studies. The water in the pipe of seesaw wells was pumped out and water samples were collected in plastic bottles washed with acid. Acids were added to the samples for preservation and transported to the Laboratory for analysis.

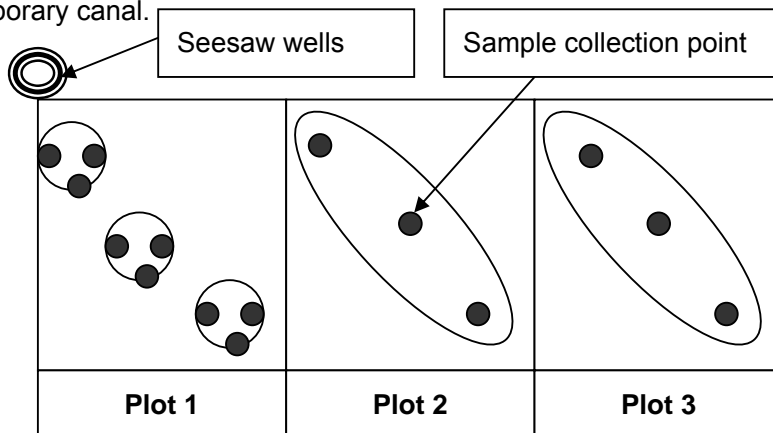
There after, 10 seesaw wells were selected for further studies from the three Village Development Committees (VDCs). Their depths ranges from 5 to 22 m, with arsenic contents ranging from < 0.005 to 1.014 mg L<sup>-1</sup> (Table 3.2.1). The groundwater from these wells is mostly used for irrigation from January to May.

Arsenic content in irrigation water (mg L <sup>-1</sup> )	No. of seesaw wells
0.00 – 0.01	3
0.15 – 0.25	3
0.50 – 0.60	2
0.80 – 1.00	2

**Table 3.2.1. Distribution of selected seesaw wells with reference to arsenic content in water**

### 3.2.2 Selection of soil plots and soil sample collection

To standardize the sample collection process, an area of 465 m<sup>2</sup> (approx.) around each well was detected as one plot, which was then subdivided into three subplots (Fig. 3.2.2). The subplots were separated by small soil dams (bund). The plots were irrigated through a small temporary canal.



**Fig. 3.2.2. Design of plots and soil and plant samples collection scheme**

Composite soil samples were collected at two different depth 0-10 cm. and 10 -20 cm, which is considered as the major layers of plant root activity in terms of nutrient uptake (Roberts et al., 1997). The general characters of soils are presented in Table 3.2.2.

Soil code	pH (1:2.5)		Texture			OM %	Oxalate extractable		Phosphorous		CEC (meq kg <sup>-1</sup> )
	H <sub>2</sub> O	CaCl <sub>2</sub>	Clay %	Silt %	Sand %		Fe (ppm)	Al (ppm)	TP (ppm)	AP (ppm)	
0	8.4	7.9	6.8	35.8	57.4	1.7	1942	11.3	898	246	70
1	8.3	7.9	8.3	36.2	55.5	1.9	3882	10.5	472	90	90
2	8.4	7.9	8.2	36.4	55.4	1.9	1073	11.0	608	191	100
3	8.4	7.8	6.7	38.1	55.3	1.3	2521	11.3	877	135	80
4	8.5	7.8	6.6	35.8	57.6	1.3	1926	10.3	800	200	80
5	8.4	7.9	8.5	36.6	54.9	1.7	1661	10.4	380	79	100
6	8.1	7.6	10.0	37.4	52.6	2.2	870	11.3	824	366	110
7	8.5	7.9	6.6	34.3	59.2	1.6	1970	10.4	501	254	90
8	8.5	7.9	6.7	34.6	58.7	1.6	1626	10.3	500	255	90
9	8.4	7.9	8.6	36.4	65.0	1.9	2295	10.5	752	189	100

**Table 3.2.2. General characterization of the soils**

*TP = Total Phosphorus, AP = Available phosphorus*

### 3.2.3 Selection of plants and sample collection

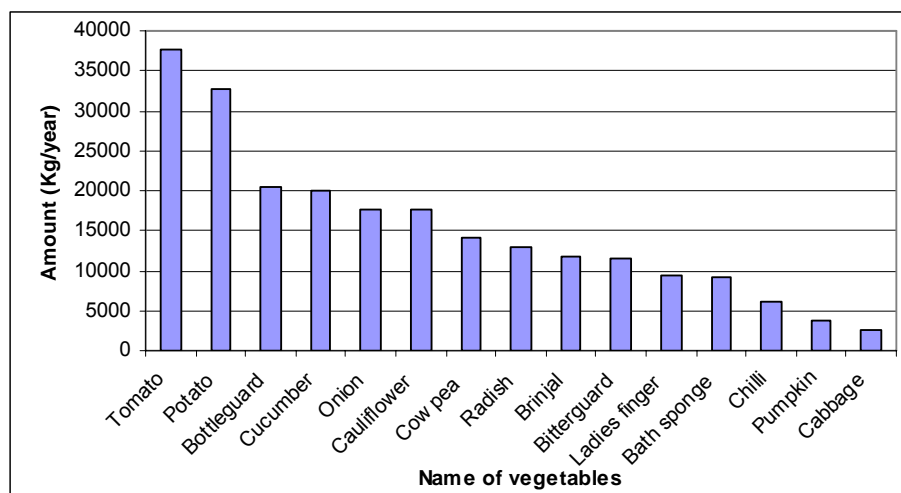
Informations were collected by interviewing the farmers. This survey was done to estimate the amount of production of vegetables, the areas of field irrigating by groundwater and the amount of water used for irrigation during each season of vegetable plantation. At average, 20 types of vegetables and 2 crops are cultivated in one year (Table 3.2.3). Generally, farmers harvest the vegetables in the winter and spring season and rice in the rainy season. Some times farmers cultivate wheat or maize also in spring season.



S.No.	Nepali Name	English Name	Scientific Name	S.No.	Nepali Name	English Name	Scientific Name
1	Bodi	Cow pea	<i>Vigna angulata</i>	12	Ghiraula	Bath sponge	<i>Luffa cylindrica</i>
2	Pyaz	Onion	<i>Allium cepa</i>	13	Pharsi	Pumpkin	<i>Cucurbita pepo</i>
3	Aalu	Potato	<i>Solanum tuberosum</i>	14	Kharbuja	Watermelon	<i>Citrullus lanatus</i>
4	Karela	Bitterguard	<i>Momordica charantia</i>	15	Bhindi	Ladies finger	<i>Hibiscus esculentus</i>
5	Khursani	Chilli	<i>Capsicum annum</i>	16	Phul Kopi	Cauliflower	<i>Brassica oleracea</i>
6	Palki sag	Spinach	*	17	Kundru	*	*
7	Lauka	Bottleguard	<i>Lagenaria siceraria</i>	18	Bhanta	Brinjal	<i>Solanum melongena</i>
8	Golbheda	Tomato	<i>Lycopersicon esculentum</i>	19	makai	Maize	<i>Maizea mays</i>
9	Banda kopi	Cabbage	<i>Brassica oleracea</i>	20	Mula	Raddish	<i>Raphanus sativus</i>
10	Kankra	Cucumber	<i>Cucumis sativus</i>	21	Tirai	*	*
11	Pidalu	Colocassia	<i>Colocassia esculenta</i>	22	Dhan	Rice	<i>Oryza sativa</i>

**Table 3.2.3. Vegetables and crops produced in Nawalparasi**

The selection of crops and vegetables was done according to the amount of production during the last years. Rice was selected among the crops because it is generally produced every year and shows high production (Fig. 3.2.3). Potato, Cauliflower, Brinjal and Onion were selected as seasonal vegetables during the study period which also produce large quantities among the varieties of vegetables. The following figure (3.2.3) shows the production of the vegetables within the study area.



**Fig. 3.2.3. Vegetable production in Manari, Devgaoun and Sukrauli of Nawalparasi district**

**(Only the vegetables which produced more than 500 Kg/year are listed in the figure.)**

The plant samples were taken from the same site or at least as close as possible to the point where the soil samples were collected. In total, 15 samples were collected from each plot for each plant. The rice and the vegetables were grown on the same sites: rice in the rainy season and vegetables in the winter and spring season, under the usual farming practices. First, the plant samples were carefully washed with arsenic free water, to remove the soil particles attached to the plant body, and then with de-ionised water. The roots, shoots, leaves and fruits were collected separately and cut into small pieces. Then the samples were air dried and packed into plastic bags.

### **3.3 Laboratory analysis**

Atomic adsorption spectroscopy coupled with a hydride generation system (HG-AAS) was used for the analysis of total arsenic. The total arsenic was measured by the reduction method where 1 ml of conc. HCl, 1 ml of a 5 % potassium iodide solution and ascorbic acid were added to 1 ml of sample, and kept overnight to make sure that the reduction of arsenate to arsenite was complete. Arsenic speciation was carried out by the ion exchange process. The Bio-Rad laboratory's AG1-X8 crystals was used as an ion exchange resin.

0.5 g of soil (dried at 105°C) and 0.2 g of plant samples (dried at 50°C) were weighed in teflon vessels. To the soil sample, 5 ml of nitric acid (65%) was added, followed by 1 ml of hydrogen peroxide (30%) and digested in a microwave oven. In the case of the plant sample, 6 ml of nitric acid (65%) was added, followed by 1 ml of perchloric acid (70%), and then 0.5 ml of hydrogen peroxide (30%). The filtrate was analyzed for total As by an Atomic Adsorption Spectrophotometer (AAS) and for total Fe by a Flame Atomic Adsorption Spectrophotometer (FAAS). Total Al and Ca were measured by ICP-AES.

### **3.4 Statistical analysis**

The data were analyzed using STATISTICA software (1984-2000) developed by Statsoft Inc., USA. An analysis of variance was performed to determine the influence of the factors such as soil depth, arsenic content and time period on the investigated variables. The p-

value was set to 0.05, to determine significant effects. To assess the differences between the individual treatments, a post-hoc comparison of means was performed using the Least Significant Difference (LSD) test.

### 4 Results and general discussions

#### 4.1 Arsenic contamination of the ground water

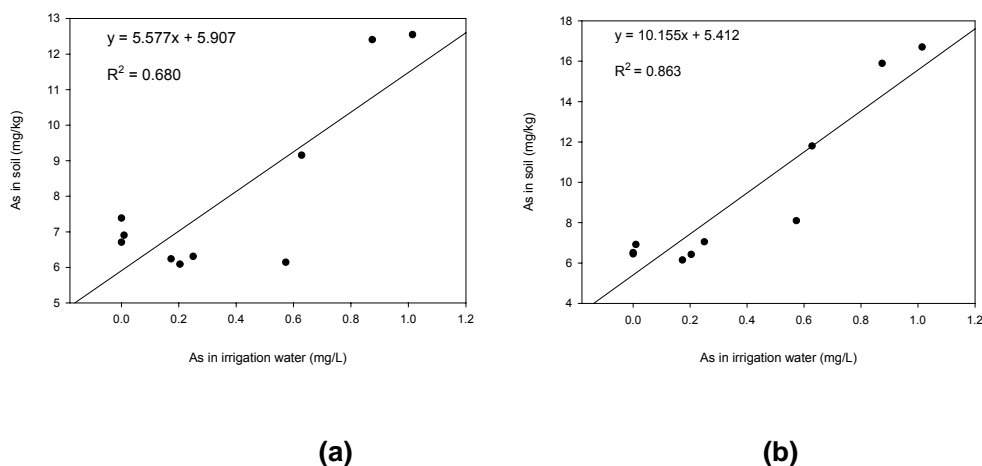
In total, 65 % of 52 wells showed an arsenic concentration higher than  $0.05 \text{ mg L}^{-1}$ , which is the Nepal interim standard for drinking water. Concentrations measured in the samples of irrigation water were ranging between  $< 0.005$  and  $1.014 \text{ mg L}^{-1}$ , with a mean concentration of  $0.14 \text{ mg L}^{-1}$ . But 36% of the pumps exceeded the FAO guideline value ( $0.1 \text{ mg L}^{-1}$ ) of As for irrigation water. When seasonal results were compared, 58% of 31 water samples collected in pre-monsoon were found lower in arsenic concentration than those of post-monsoon; in 32% of the samples, the arsenic content was increased and in 10% it remained more or less constant. The concentration of arsenite in the samples ranges from  $< 0.005$  to  $0.87 \text{ mg L}^{-1}$ , with an average concentration of  $0.11 \text{ mg L}^{-1}$ . The result showed that the percentage of arsenite is 76.3% (range: 50 – 100%).

In Bangladesh, geologically it has been supposed that arsenopyrite is the source of arsenic in the sediments varying in depth from 14 - 80 m below the surface (Hassan, 2007). This depth is subjected to oxidation and reduction of arsenopyrite due to withdrawal of water for domestic and irrigation purposes. In South and East Asia such conditions tend to occur in the shallower parts of quaternary aquifers underlying the region's large alluvial and deltaic plains e.g., Bengal basin, Irrawaddy delta, Mekong valley, Red River delta, Indus plain, Yellow River plain (The World Bank, 2004<sub>a</sub>). A survey of shallow groundwaters in Bangladesh indicates that 27% exceeded  $0.05 \text{ mg L}^{-1}$  and 46% exceeded  $0.01 \text{ mg L}^{-1}$  (BGS-DPHE, 2001). Concentrations were up to  $1.65 \text{ mg L}^{-1}$  in 206 tubewells of India tested and 57% exceeded  $0.05 \text{ mg L}^{-1}$  (Chakraborti et al., 2003). In Myanmar, 15% of groundwater samples from 8 937 tubewells exceeded  $0.05 \text{ mg L}^{-1}$  as revealed by surveys conducted in 2002 by WRUD (The World Bank, 2004<sub>b</sub>). A total of 364

samples were analyzed in Pakistan and only 2% tubewells were found arsenic contaminated with concentrations above  $0.05 \text{ mg L}^{-1}$  (Iqbal, 2001).

## 4.2 Arsenic contamination of soils

The arsenic contents in soils were measured where arsenic contaminated water was used for irrigation for 2 years. The highest arsenic content ( $16.7 \text{ mg kg}^{-1}$ ) was measured in surface soils collected from a vegetable field with a mean of  $9.2 \text{ mg kg}^{-1}$  ( $n = 50$ ), for rice field soils, it was  $12.5 \text{ mg kg}^{-1}$ , with a mean of  $8 \text{ mg kg}^{-1}$  ( $n = 50$ ). In those fields, the arsenic concentration in the irrigation water was  $1.014 \text{ mg L}^{-1}$ . Although the arsenic content in soils was positively correlated with arsenic concentration in irrigation water (Fig 4.2) the arsenic content in soils increased in the areas with  $> 0.628 \text{ mg L}^{-1}$  of arsenic in irrigation water.



**Fig. 4.2. Regression plot of As content in irrigation water Vs. soils (a) Rice field (b) Vegetable field.**

Many researchers found a positive correlation between the arsenic in irrigation water and soils (Das et al., 2004; Alam and Sattar, 2000; and others). However, the influence of arsenic concentration in irrigation water on the arsenic content of soils is controversial. In a preliminary survey of arsenic in 71 soil samples collected throughout Bangladesh it was found that in the western part of Bangladesh the highest soil concentrations ( $> 30 \text{ mg kg}^{-1}$ ) occurred, followed by the central belt, which is in agreement with groundwater

concentrations (Meharg and Rahman, 2003). Alternatively, they found low arsenic contents in the soil at various locations with high As levels in groundwater. Although they have not mentioned the reasons, there are various explanations for this phenomenon: As desorbs to the standing water and is then removed laterally; the top layer may be eroded with run off during heavy rainfall; volatilization of As during prolonged periods of flooding and leaching of standing water, desorbing and transporting As from the topsoil to deeper layers (Heikens, 2006) may be the responsible for variations of arsenic in soils.

### **4.3 Arsenic content in plants**

The mean arsenic content in rice grains (with out husk) in our study was  $0.18 \text{ mg kg}^{-1}$ . The result of our study shows the following order for average arsenic concentration in vegetables:  $0.55 \text{ mg kg}^{-1}$  in onion leaf >  $0.45 \text{ mg kg}^{-1}$  in onion bulb with skin >  $0.33 \text{ mg kg}^{-1}$  in cauliflower >  $0.09 \text{ mg kg}^{-1}$  in brinjal > not detected in potato with skin. Significant correlations are obtained between arsenic in irrigation water and in the root of brinjal, followed by potato, cauliflower and rice. But the correlation between water vs. leaf of brinjal is followed by cauliflower, onion, rice and potato. Similarly, the correlation between arsenic in water and the edible parts are calculated as with onion ( $r^2 = 0.89$ ), brinjal ( $r^2 = 0.59$ ), rice ( $r^2 = 0.38$ ) and with cauliflower ( $r^2 = 0.19$ ). A positive correlation was found between As in the groundwater and As in the rice collected throughout Bangladesh (Williams et al., 2006).

A study conducted in Bangladesh found that the rice grain contained  $0.7 \pm 0.2 \text{ mg kg}^{-1}$  and  $0.6 \pm 0.0 \text{ mg kg}^{-1}$  in two different varieties of rice BRRI 1 and BRRI 28 respectively in the areas with  $0.07 \text{ mg L}^{-1}$  of arsenic in irrigation water (Rahman et al., 2007). In another study, it was reported  $0.42 \text{ mg kg}^{-1}$  arsenic rice grain when  $8.0 \text{ mg L}^{-1}$  of arsenic contaminated water was irrigated (Abedin et al., 2002<sub>a</sub>). The arsenic content of vegetables from the UK was approximately 2 to 3-fold lower than those observed for vegetables imported from Bangladesh (Ramlli et al., 2005). There is no standard maximum level of arsenic in food in South and East Asian countries (The World Bank, 2004<sub>b</sub>).

Arsenic accumulation and translocation factors in plants were calculated by using the following relationship:

$$\text{As accumulation factor} = \text{total As in plant parts (mg kg}^{-1}\text{)} / \text{total As in soil (mg kg}^{-1}\text{)}$$

$$\text{As translocation factor} = \text{total As in aerial plant parts (mg kg}^{-1}\text{)} / \text{total As in the roots (mg kg}^{-1}\text{)}$$

The rice roots showed the highest accumulation factor (2.43), compared with arsenic in the irrigation water, with 0.628 mg L<sup>-1</sup> (Table 4.3.1). The arsenic accumulation factors for vegetable roots were found very low (< 0.5) in comparison with rice roots (> 1). The highest accumulation factors in all samples of rice and vegetables were observed in roots, followed by shoots > leaves > edible parts. A similar result was found by Abedin et al. (2002<sub>a</sub>) and Abedin et al. (2002<sub>b</sub>) in pot experiments with rice plants. The highest accumulation factor among the vegetables was obtained for cauliflower roots (0.33) compared with an arsenic concentration of 0.250 mg L<sup>-1</sup> in the water (Table 4.3.2). When arsenic accumulation factors of edible parts were compared, the highest accumulation factor was found in cauliflower, followed by onion > rice > brinjal > potato (Table 4.3.1 and 4.3.2).

As in irrigation water(mg/L)	As in soil (mg/kg)	Accumulation factor				Translocation factor		
		Root	Shoot	Leaf	Grain	Shoot	Leaf	Grain
<0.005	7.4	1.09	0.16	0.17	0.01	0.15	0.16	0.01
0.173	6.2	1.50	0.27	0.25	0.01	0.18	0.17	0.01
0.250	6.3	2.06	0.54	0.50	0.04	0.26	0.24	0.02
0.628	9.1	2.43	0.45	0.39	0.04	0.19	0.16	0.01
1.014	12.5	1.53	0.25	0.24	0.02	0.17	0.16	0.01

**Table 4.3.1 Arsenic accumulation and translocation factor for rice (dry matter basis).**

The mean arsenic accumulation factors for roots, shoots, leaves and grains of rice were 1.72, 0.35, 0.31 and 0.01, respectively (Table 4.3.1). This indicates its strong ability to accumulate arsenic in the plant body, similar to some *Pteris Taxa* plants (Wang et al., 2007). But the translocation factor for shoots, leaves and grains of rice was very low (<

0.2) in comparison with the accumulation factor (Table 4.3.1). In turn, the high amounts of arsenic accumulated in rice roots will remain in the soil while other parts are removed during harvesting. Thus, the arsenic content of contaminated roots will be an additional source of arsenic accumulation in the soils. The accumulation factors in all studied vegetables were lower than in rice, although they followed the same pattern as rice, i.e. roots showed the highest accumulation factor and edible parts showed the lowest one, except in onions (Table 4.3.2). The onion leaves showed a higher accumulation factor than its roots and bulb. In general, the plants which are not hyperaccumulators of arsenic have accumulation factors ranging from 0.01 - 0.1 (Kloke et al., 1984). Similarly, in this study the accumulation factor between the soil and the edible parts of all plants was < 0.1 (Table 4.3.1 and 4.3.2), which has also been reported by Warren et al. (2003) for some vegetables grown in untreated arsenic contaminated sites in the UK. The translocation factor to edible parts in general is higher in vegetables (> 0.1) than in rice, except for potatoes (Table 4.3.3).

As in irrigation water(mg/L)	As in soil (mg/kg)	Potato				Cauliflower				Onion			Brinjal			
		Root	Shoot	Leaf	Fruit	Root	Shoot	Leaf	Fruit	Root	Leaf	Fruit	Root	Shoot	Leaf	Fruit
<0.005	6.4	0.03	0.01	0.01	ns	0.16	0.09	0.03	0.01	ns	0.01	ns	0.05	0.02	ns	ns
0.173	6.1	0.03	0.01	0.01	ns	0.27	0.12	0.05	0.03	ns	0.01	ns	na	na	na	na
0.250	7.0	0.04	0.02	0.01	ns	0.33	0.12	0.05	0.09	0.01	0.04	0.03	0.07	0.03	0.02	0.02
0.628	11.8	0.04	0.02	0.01	ns	0.29	0.10	0.07	0.03	0.02	0.10	0.08	na	na	na	na
1.014	16.7	0.05	0.01	0.01	ns	0.19	0.08	0.05	0.02	0.02	0.07	0.06	0.07	0.03	0.01	0.01

**Table 4.3.2 Arsenic accumulation factor for vegetables (dry matter basis).**

*na = not available, ns = not significant*

Higher translocation factors are calculated for vegetables (> 0.3) than for rice (< 0.3) (Table 4.3.3). The translocation factors follow the same trends as the accumulation factors, i.e. in decreasing order from shoot to leaves and to edible parts. The highest translocation factors were observed in onion leaves, and the lowest in brinjal leaves. Similarly, the translocation factor in the edible part of onions was the highest, but the translocation factor was not calculated for potatoes because of their very low arsenic



content. However, the Concentrations in all plant parts increased with the exposure concentration (Abedin et al., 2002<sub>a</sub>; Abedin et al., 2002<sub>b</sub>).

As irrigation water(mg/L)	As in soil (mg/kg)	Potato				Cauliflower				Onion			Brinjal			
		Root	Shoot	Leaf	Fruit	Root	Shoot	Leaf	Fruit	Root	Leaf	Fruit	Root	Shoot	Leaf	Fruit
<0.005	6.4	0.03	0.01	0.01	ns	0.16	0.09	0.03	0.01	ns	0.01	ns	0.05	0.02	ns	ns
0.173	6.1	0.03	0.01	0.01	ns	0.27	0.12	0.05	0.03	ns	0.01	ns	na	na	na	na
0.250	7.0	0.04	0.02	0.01	ns	0.33	0.12	0.05	0.09	0.01	0.04	0.03	0.07	0.03	0.02	0.02
0.628	11.8	0.04	0.02	0.01	ns	0.29	0.10	0.07	0.03	0.02	0.10	0.08	na	na	na	na
1.014	16.7	0.05	0.01	0.01	ns	0.19	0.08	0.05	0.02	0.02	0.07	0.06	0.07	0.03	0.01	0.01

**Table 4.3.3 Arsenic translocation factor for vegetables (dry matter basis).**

*na = not available, ns = not significant*

#### **4.4 Arsenic distribution in soil aggregate fractions**

Significant differences ( $p < 0.05$ ) in the load of arsenic were observed between soils of the rice field and those of the vegetable field, indicating that both, the kind of plant cropping and the aggregate distribution clearly influence the arsenic content in soil. The load of As, Fe, Mn, Al, and Ca was found higher in smaller aggregate sizes. Arsenic sorption in soils is primarily related to the content of Fe and Al oxides and other elements (Fayiga et al., 2007; Mahimairaja et al. 2005; Wilkie and Hering, 1996, Fuller et al., 1993). This may be the reason for the positive correlation of arsenic with Fe, Mn, Al and Ca present in the smallest aggregate fractions. Arsenic showed a moderate correlation with Fe and Mn in smallest fraction of rice field soils but it showed the strongest correlation with Ca in the smallest fraction of vegetable field soils compared with Fe, Mn, and Al.

#### **4.5 Role of Redox potential and DAP fertilizer on de-sorption of As from soil**

The behaviour of As is distinctly different under non-flooded (aerobic) and flooded (anaerobic) soil conditions. Therefore, laboratory experiments were performed to know the role of phosphorus on de-sorption of arsenic under aerobic and anaerobic conditions.

After the addition of Di-Ammonium Phosphate (DAP) fertilizer, the change in Eh is found more or less similar to the experiment without DAP fertilizer treatment in the case of alkaline soils. But a strongly reducing condition ( $Eh = -500$  mV) was achieved in acidic soils only after addition of DAP fertilizer. In alkaline soils, the pH was found decreased with the decrease of Eh values but not lowered like in experiment sets without treatment of DAP. While in acidic soils, pH values steeply increases from 3.9 to 7.6 and 3.9 to 7.9, respectively, at first then slightly lowered. Therefore it can be concluded the soils maintain nearly neutral pH values either by increase or decrease of pH after addition of DAP fertilizer.

Soluble As increased about 5 times after the addition of DAP fertilizer to both, acidic and alkaline soils although the soluble Fe and Ca were not increased. After the addition of DAP, the soluble As was not decreasing in highly anaerobic conditions like in the soil solution without DAP treatment. This might be a reason that high amounts of As are released due to the addition of DAP and sulphide ions are insufficient to produce insoluble compounds with As. An decrease in pH increases the P binding capacity of Fe, due to ligands exchange reactions where phosphate ions replace OH (Boström et al., 1982). This could be a reason for the decrease of soluble Fe after the addition of DAP. Dissolved As concentrations increased during the same time when Fe dissolution was occurring (Reynolds et.al, 1999). But it was found consistent only when there was not addition of DAP fertilizer. It is interesting that soluble Ca was found lower than Fe after addition of DAP fertilizer in case of alkaline soils. At higher pH, the P binding capacity of Ca is high ((Boström et al., 1982). This indicates the phosphate ions at first replaced As ions associated with Ca than Fe at a higher pH. When the pH started to decrease the phosphate replaced the As ions associated with Fe.

### 5 Conclusions

This study investigates the current scenario with regard to the occurrence and distribution of arsenic contamination in three villages of the Nawalparasi district. The irrigation water in the study area is highly contaminated, with a mean arsenic concentration of  $0.14 \text{ mg L}^{-1}$ . The mean addition of arsenic to soil by irrigation with ground water is  $1.50 \text{ mg kg}^{-1} \text{ year}^{-1}$ . But the arsenic addition can reach up to  $10.19 \text{ mg kg}^{-1} \text{ year}^{-1}$ , indicating that continuous use of contaminated water makes the soil rich in arsenic within decades. Therefore it can be concluded that although there are several reasons for arsenic contamination in soils, the pathway through irrigation water should not be neglected in the study areas. The irrigation water is rich in arsenite (mean concentration =  $0.11 \text{ mg L}^{-1}$ ) so that there is also a high probability of arsenic uptake by the plants grown on the soil.

The correlation between the arsenic content in plants and the arsenic concentration of the irrigation water was found better than between the arsenic content of plants and soils. This indicates that the uptake of arsenic by plants is mainly influenced by the amount of arsenic contained in irrigation water. The continuous use of arsenic contaminated water higher than  $0.250 \text{ mg L}^{-1}$ , for irrigation for longer period increases the risk of arsenic contamination in the food chain.

Our study showed that the distribution of soil aggregate fractions depends on the kind of cropping systems which have different distribution loads of As. As contained in smaller aggregate fractions ( $\leq 0.63 \text{ mm}$ ) showed strong correlation with Fe, Mn and Ca only in case of the vegetable cropping field. In all aggregate fractions, Al showed no clear relation with arsenic, neither in rice fields and nor in vegetable fields.

The As solubility was increased by about 5 times after the addition of Di-Ammonium Phosphate (DAP) fertilizer under all redox potential conditions (aerobic to anaerobic) while

soluble Fe and Ca was not increased. This shows that the addition of DAP fertilizer is highly influential on the increase of soluble As in alkaline and acidic soils through ion exchange mechanisms although the redox potential and pH values of the soils also control the solubility of As through dissolution of Fe and Ca.

This research focused only on highly arsenic contaminated areas. However, a national survey is required in the future to get a detailed picture of arsenic contamination of irrigation water. Further research is also needed to assess the contamination of soil, crops and vegetables in order to analyze possible impacts on public health.

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## 9 Annexes

**Annex-1: Screening of arsenic in irrigation water used for vegetable production in Nepal.** *Archives of Agronomy and Soil Science*, 54(01), pp. 41 - 51. DOI: 10.1080/03650340701628197

Dahal B.M., Fuerhacker M., Mentler A., Shrestha R.R. & Blum W. E. H. (2008).

**Annex-2: Arsenic contamination of soils and agricultural plants through irrigation water in Nepal.** *Environmental Pollution* 155 pp.157-163.

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**Annex-3: Arsenic distribution in soil aggregate fractions two different cropping fields in Nepal. Accepted by:** *Die Bodenkultur Journal for Land Management, Food and Environment*

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**Annex-4: Arsenic solubility in soil as affected by redox conditions and phosphorus fertilizer application.** Submitted to: *Geoderma*

Dahal B.M., Mentler A., & Blum W. E. H. (2008).

*Note: See papers in hardcopy of thesis*

## 10 Table of abbreviations

AAS	Atomic Adsorption Spectrometry
Al	Aluminium
As	Arsenic
Ca	Calcium
BGS	British Geological Society
DAP	Di-Ammonium Phosphate
DPHE	Department of Public Health Engineering, Bangladesh
DW	Dry Weight
DWSS	Department of Water Supply and sewerage
ENPHO	Environmental and Public Health Organization
FAO	Food and Agriculture Organization, United Nations
Fe	Iron
HG-AAS	Hydride Generators-Atomic Adsorption Spectrophotometer
F-AAS	Flame Atomic Adsorption Spectrophotometer
ICP-AES	Inductive Coupled Plasma-Atomic Emission Spectrophotometer
MDG	Millennium Development Goal
Mn	Manganese
MOFSC	Ministry of Forest and Soil Conservation, Nepal
NAS	National Academy of Sciences, Washington DC
NASC	National Arsenic Steering Committee, Nepal
ORP	Oxidation Reduction Potential
P	Phosphorus
WHO	World Health Organization
WRUD	Water Resources Utilization Department, Myanmar

## 11 Units of measurement

ng m<sup>-3</sup>      nanograms per cubicmeter

µg            microgram

mg            milligram

kg            kilogram

L             liter

µg L<sup>-1</sup>        micrograms per liter

mg L<sup>-1</sup>        milligrams per liter

mg kg<sup>-1</sup>       milligrams per kilogram

m             meter

km            kilometer

## 12 Curriculum Vitae

### Personal data

Name	Binod Mani Dahal
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