Master's Thesis – master Water Science and Management

A study on the primary sources of natural arsenic pollution in the Ganges and Indian Brahmaputra River Basins

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Abstract

Arsenic, a pervasive and naturally occurring trace metal in the earth's crust, is considered to be highly toxic and poses serious health issues. Millions of people are threatened by arsenic polluted groundwater resources, especially in the Southeast Asian densely populated river deltas like the Ganges-Brahmaputra-Meghna (GBM) delta. Although the fate and mobilization of arsenic in groundwater have been largely studied and understood, there are still many uncertainties about the primary source of arsenic. Therefore, this study aimed to identify the primary arsenic sources which cause the groundwater pollution in the downstream deltaic areas of the Ganges and Brahmaputra River Basins (GRB and BRB). The GRB is an interesting research area as its Gangetic Plain acts as the connection between the Himalayas and the Bengal delta, therefore making the image complete of the arsenic route from geological source to sink. The general setup of this research consisted of a data mining exercise whereby as many relevant literature sources as possible were used in order to construct a regional geographic image of the arsenic groundwater distribution in the research area. The first and second step of the methodology consisted of collecting data on groundwater concentrations of arsenic and heavy metals (associated with sulphides) for the GRB and Brahmaputra river basin (BRB). This was followed by the third step whereby the distribution of elevated arsenic groundwater concentrations was compared with the heavy metal anomalies distribution and linked to local surface geology. The final step consisted of indicating possible arsenic sources by connecting features of the surface geology with the mobilisation of (groundwater) arsenic. The general results showed that both the GRB and Indian BRB are affected by high arsenic groundwater concentrations, whereby it is pointed out that arsenic groundwater anomalies are also found within the Himalayas up to altitudes around 3500 m.a.s.l and not only in the Terai, Gangetic Plains and GBM delta. The results indicated that there were no clear correlations between the occurrence of high concentrations of the heavy metals and the distribution of arsenic groundwater anomalies. In the Nepalese Himalayas, the Seti, Ranimatta and Ulleri Formations seem to be acting as a source of arsenic release, whereby the weathering of surface rocks containing biotite and muscovite is a mechanism that possibly contributes significantly to local arsenic-enriched groundwater. Further results of this study pointed towards the leucogranites of the Tethyan Himalayas as being a likely primary source of arsenic. Weathering of these rocks appears to be causing arsenic groundwater enrichment in some high-altitude and remote areas (in particular Mustang Valley) in Nepal. However, the leucogranites are also present in areas with only low arsenic groundwater concentrations. Hence, they do not always act as a source for elevated arsenic groundwater concentrations throughout the research area. Moreover, results indicated that regions with an absence of leucogranites, such as in the Indian states of Nagaland and Manipur, also appear to be able to have high arsenic groundwater concentrations. Thus, the leucogranites are not the single source for elevated arsenic groundwater concentrations and other (local) yet-unknown arsenic sources need to be considered as well.

Source image on front page: UNICEF & WHO., (2018). Arsenic Primer: Guidance on the Investigation & Mitigation of Arsenic Contamination.

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1. Introduction

The arsenic crisis

Arsenic, a pervasive and naturally occurring trace metal in the earth's crust, is considered to be highly toxic and is seen as one of the 20 most hazardous substances (ATSDR, 2017; Guo et al., 2014). Millions of people are threatened by arsenic polluted groundwater resources, especially in the Southeast Asian densely populated river deltas like the Ganges-Brahmaputra-Meghna (GBM) delta. In particular the young, Quaternary sediments which compose the major deltaic and alluvial plains and inland basins of Southeast Asia, are prone to promote the arsenic groundwater problem (Smedley, 2003). Besides the aquifers in Southeast Asia, groundwater pollution with arsenic has been detected in many other parts of the world including Argentina, Chile, China, Ghana, New Zealand, Russia, UK, Hungary and Taiwan (Chakraborti et al., 2002). In continental Asia, critically high concentrations are found in the Bengal basin of eastern India and Bangladesh (Safiullah, 2006; Chakraborti et al., 2001), the lowland region of the Terai in Nepal (Tandukar et al., 2006), the Red River delta in Vietnam (Berg et al., 2001), the Yellow River plain and additionally some northern Chinese basins (Guo et al., 2001). These regions are mostly flat-lying fertile plains and unfortunately are often densely populated. Since groundwater is the main source of drinking water in these areas, a large number of people is significantly impacted by a decrease in groundwater quality due to arsenic pollution (Tareq et al., 2010).

Arsenic poses serious health issues as it is proven that organisms are harmed by the exposure of sufficiently high arsenic concentrations in soil, sediments and water. Human beings are exposed to arsenic ingestion (arsenic poisoning or "arsenicosis") through polluted drinking water, as well as consuming food and also inhalation of air (Tareq et al., 2010). Intake of high levels of arsenic can lead to various health problems such as skin malignancy; gastrointestinal uneasiness; dermal hyperkeratosis and also cancer (Morton & Dunette, 1994). It is believed that currently about 200 million people are endangered by elevated arsenic groundwater concentrations that exceed the acceptable guideline of arsenic in drinking water of 10 µg/L, as stated by the World Health Organization (WHO) (Mukherjee et al., 2009). Moreover, researchers of the WHO have described the current arsenic pollution in Bangladesh and India as "the largest mass poisoning of a population in history" (WHO, 2000). Although the WHO has lowered the threshold for acceptable arsenic concentration in drinking water from 50 µg/L to 10 µg/L in order to raise awareness of the health concerns related to the element, some countries including India and Bangladesh still maintain a threshold value of 50 µg/L (Tareq et al., 2010).

Besides the physical impacts, victims from arsenic poisoning also face psychological effects as they are often mistreated and forced to become social outcasts or misfits, rather than being pitied or nurtured by their communities. Since arsenicosis is a relatively new phenomenon in Southeast Asia, it is still poorly understood, especially among the population in rural areas (Hassan et al., 2005). Because of this lack of knowledge, arsenic patients are often depicted as 'dangerous' people and are stripped of their societal status. This has led to a serious problem of social instability in arsenic affected areas. Victims have become isolated from their families, face divorce, unemployment, domestic violence, discrimination and even physical torture (Argos et al., 2007).

Arsenic fate, mobilisation and sources

Since the first large occurrence of arsenic in well water was identified in the 1990s, great attention has been paid towards investigating the fate, mobilisation and sources of arsenic in the environment

(Anawar et al., 2003; Parrone et al., 2020). The most relevant mechanism responsible for the occurrence of groundwater arsenic is the water-rock interaction paired with favourable biogeochemical conditions (Mukherjee et al., 2014). The mobilisation of arsenic is thought to be caused by volcanism, biochemical activities and chemical and physical weathering (Mondal et al., 2010; Guo et al., 2017). Many hypotheses have been proposed about the exact mechanisms responsible for the mobilisation of arsenic in groundwater environment. The most widely accepted and used hypotheses include: (1) oxidation of pyrite; (2) competitive ion exchange; (3) reductive dissolution of iron oxyhydroxides; (4) mobilisation of arsenic caused by self-organizing geochemical processes in deltaic sedimentary environments (Tareq et al., 2010).

High arsenic levels observed in groundwater can result from either human activities or natural processes. Arsenic may be anthropogenically introduced to the environment as it is present in acid mine drainage, wood preservatives, fertilizers and various other sources such as herbicides, semi-conductors and pharmaceuticals (Jayasumana et al., 2015; Chen & Olsen, 2016). In addition, cement manufacturing, paper production, wastes and fossil fuels also contribute to anthropogenic arsenic contamination (USEPA, 2002).

Despite naturally occurring, the abundance of arsenic in the earth's continental crust is not high. Arsenic is naturally present in some rocks, whereby the arsenic is released into the environment after weathering and erosion of the rocks. Other natural arsenic sources include volcanic activities (e.g. Tedd et al., 2017). Physical, chemical and microbiological weathering commonly mobilise arsenic from arsenic containing minerals. Sulphide minerals (mainly pyrite) and iron oxides are the most common sources of arsenic discharge (Kumar & Singh, 2020). Moreover, it is also documented that Holocene alluvial sediments with slow hydrogeological flow rates, organic-rich or black shales, geothermal activity (geothermal springs) and coal all contribute significantly to high arsenic concentrations in groundwater (Smedley & Kinniburgh, 2002; Shaji et al., 2021).

Although the fate and mobilization of arsenic in groundwater have been greatly studied and understood, up till now, there are still many uncertainties about the primary source of arsenic. However, it is universally accepted that arsenic has a geogenic source and high concentrations of arsenic in groundwater in Southeast Asia are coupled to the natural weathering of the Himalayan belt (e.g. Gurung et al., 2005; Guillot & Charlet, 2007; Guillot et al., 2015; Mueller, 2017). The Himalayan foreland basin and the Bengal delta, considered as globally one of the largest modern day fluvial deltas, is build up by arsenic-laden sediments which were carried by the Ganga-Brahmaputra River system (France-Lanord et al., 1993; Garzanti et al., 2004). Estimates of the amount of sediments that is transported by the Ganga-Brahmaputra Rivers from the Himalayan range to the fluvial delta is at about 1800 tonnes/km^2 of the catchment area, whereby estimates of suspended matter discharge range between 540 to 1175 million tonnes/year (Milliman & Sivitsli, 1992).

Pinpointing the primary source of arsenic is rather troubling, as stated by Guilliot & Charlet (2007): "one of the main problems to depict the source of arsenic is that this element is very mobile and can be easily removed and recombined from the source during alteration processes, transport and mobilization in sediments". Some studies (e.g. Saunders et al., 2005) proposed that the first foothills of the Himalayas (i.e., the Siwalik Group), are the most probable provenance of arsenic. However, the theory of the Siwaliks being the primary arsenic provenance might only be a secondary sink at most. This was indicated by Mukherjee er al. (2014), who suggested that the main source of high arsenic in groundwater in southeast Asia should be found further north in the Himalaya.

Considering the primary and secondary provenance of arsenic, there are two different main theories in the literature:

- The original source of arsenic is the Qamdo-Simao (QS) volcanic and ophiolite province, based north of the Namche Barwa syntaxis, close to the Indo-Myanmar border and arsenic was transported toward the Siwalik foreland basin during the Miocene (Stanger, 2005).
- The original source of arsenic are the ophiolites found in the Indus-Tsangpo suture zone, which before being removed by extensive weathering during the Holocene, supplied the Siwaliks with sediment between the Miocene and Pleistocene (Guilliot & Charlet, 2007).

Although these two theories are the most widely accepted ones, a number of other potential arsenic sources is listed in the literature (Mukherjee et al., 2006; Mukherjee et al., 2009):

- The Gondwana coal in the Rajmahal Traps and the overlying basaltic rocks.
- The Bihar mica-belt.
- The isolated sulphide outcrops in the Darjeeling Himalayas.
- The Gorubathan base-metal deposits in the eastern Himalayas.
- Metapelites and leucogranites in the higher Himalayas (Mueller, 2018).
- The black schists from the Lesser Himalaya (Guillot et al., 2015).

The locations of these (potential) arsenic sources are presented in Appendix A1.

Research aim

Since there are still many uncertainties about the specific primary source of arsenic in Southeast Asia, this study aims to find the primary arsenic sources which cause the groundwater pollution in the downstream deltaic areas. In order to achieve this, an attempt is made to map the arsenic concentrations in the Ganges River Basin (GRB), including the adjacent Himalayan foreland and higher mountain ranges. The GRB is an interesting research area as its Gangetic Plains act as the connection between the Himalayas and the Bengal delta, therefore making the image complete of the arsenic route from source to sink. Furthermore, this area has witnessed an extensive amount of research concerning arsenic contamination since groundwater arsenic was first discovered to exceed the drinking limit of 50 µg/L back in 2002 (Chakraborty et al., 2003). Studies of the last two decades pointed out that the eastern part of the Gangetic Plains, mainly the Middle Gangetic Plain (MGP), is particularly affected by arsenic pollution (Saha & Sahu, 2016). Therefore, an investigation is needed about the primary sources of the arsenic that cause the contamination and hence threaten the shallow aquifer-based drinking water supply. Such a study helps us to better understand the occurrence and behaviour of arsenic in the environment and can be helpful for communities in guiding towards safer drinking water sources.

The main research question is: *What is the primary source of arsenic in the Ganges and Indian Brahmaputra River Basins?* This research question is divided into the following sub-questions:

- Where are arsenic anomalies, within the research area as based on groundwater arsenic concentrations?
- Where are areas located with only low arsenic groundwater concentrations (<10 µg/L)?
- Can the occurrence of heavy metal anomalies, associated with sulphides (such as Fe, Cu, Pb, Co, Zn and Ag), be used as proxy for the source of arsenic?
- Are the arsenic anomalies (and associated heavy metal concentrations) linked to primary arsenic sources?

The following hypotheses constructed from the literature will be tested:

1) The primary source of arsenic is in the Qamdo-Simao volcanic and ophiolite province.

2) The primary source of arsenic are the ophiolites of the Indus-Tsangpo suture zone.
3) The primary source of arsenic is in another area (e.g. leucogranites).

2. Study area & background information

Since the literature has guided the search for the primary source of arsenic to be found in the Himalayas, this chapter will give a short overview of the geology of the Himalayan range, followed by an description of the research area and lastly, a brief elaboration of the hypotheses found in the literature.

Himalayan geology

The Himalayas are a mountain range, located in continental Asia and separate the Indian subcontinental plain from the Tibetan Platea. The Himalayas are home to some of the tallest mountain peaks in the world. Regarding the geology, the Himalayas can be generally categorized into 4 different tectonic units. When looking at an area where all four major Himalayan tectonic units are extensively exposed, the Narayani basin in Nepal is a good example (figure 1): (1) located at the base of the South Tibetan Detachment system (STDS) is the Tethys Himalaya; (2) situated at the base of the Main Central Thrust I (MCT I) are the Higher Himalayan Crystallines (HHC); (3) at the base of the Main Boundary Thrust (MBT), is the Lesser Himalaya (LH), which is divided into the lower and upper Lesser Himalaya; and (4) at the Main Frontal Thrust (MFT) is the Siwaliks and the Quaternary foreland basin (Gurung et al., 2005; Guillot, 1999). These units have a wide variety of different igneous, sedimentary and metamorphic rocks. The differential erosion of this wide variety of rocks accounts to some of the groundwater arsenic heterogeneity in the foreland and delta (Shah, 2008).

The Tethys Himalaya unit consists of 10 km of different metasedimentary rocks (shales, quartzites, limestones and calc-shists) which range from Cambrian to Jurassic (Colchen et al., 1986). Within the Tethyan rocks is the Manaslu leucogranite (Guillot et al., 1995). The Higher Himalayan

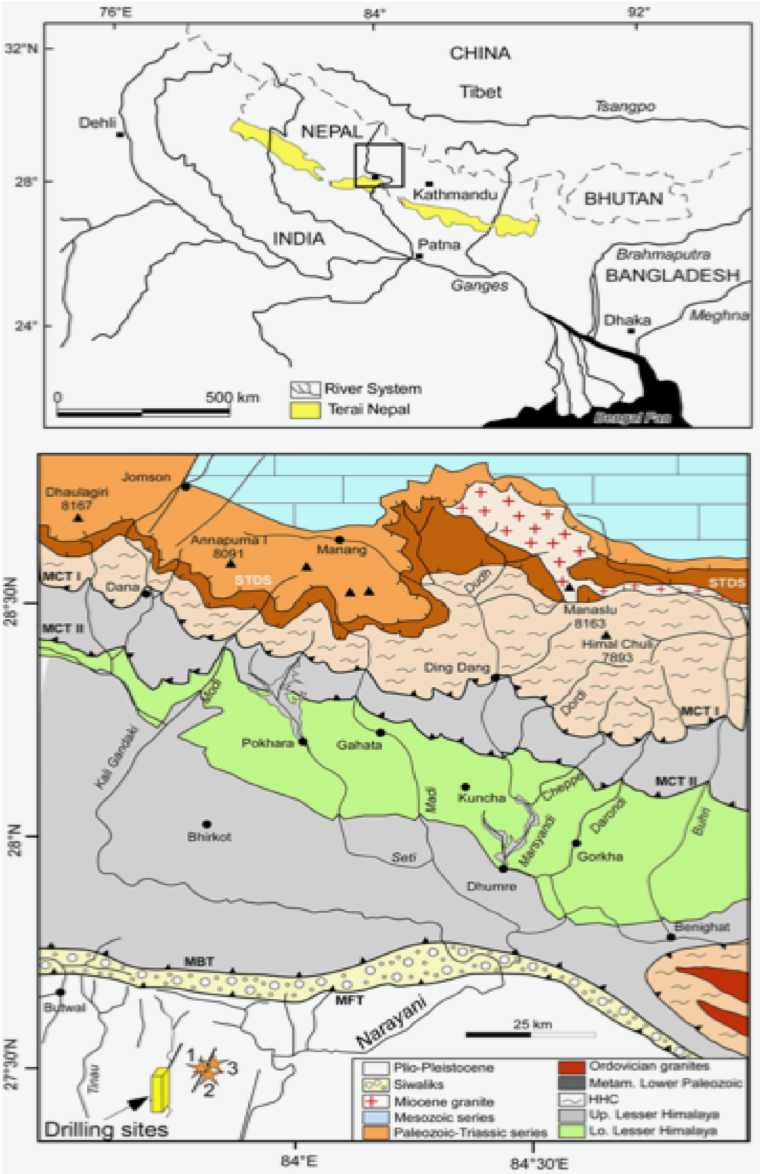

Figure 1: The Ganga-Brahmaputra watershed with the location of the Terai in Nepal and a geological map of the Himalayan range in Nepal (Guillot et al., 2015).

Crystallines can be regarded as a metamorphic unit with 2-10 km thick paragneisses, 3 km thick gneisses with calc silicate minerals and 300-500 m thick orthogneisses which are metamorphosed granite from the Lower Paleozoic (Colchen et al., 1986). The Lesser Himalaya includes mostly unfossiliferous metasediments which consist of quartzites and phyllites (Kuncha Group). Overlying this group are dolomitic meta-carbonates with aluminium-rich schists, quartzites, and dominant black schists (Colchen et al., 1986). The Cenozoic foreland basin of the Himalayan belt is represented by the Siwaliks, which have a local thickness of 6 km in Nepal (Huyghe et al., 2005). A typical coarsening-upward succession can be observed in the three units of the Siwaliks. These units consist of fluvial channel sandstones (lower unit), very thick channel sandstones (middle unit) and gravelly braided river conglomerates (upper unit) (Mugnier et al., 1999). At the foot of the Siwaliks lies a remarkably flat plain known as the Terai at between 60 and 360 m above sea level. In geological terms, the Terai is an active foreland basin which is constructed of Quaternary sediments that mostly include silt, sand, gravel and clay. Sediments are transported into the Terai by many rivers that flow southward from the Himalayan mountain range (figure 1) whereby minor rivers originate from the adjacent Siwalik Hills (Shukla & Bora, 2009).

Research area

During this study, strong emphasis was put on the initial research area of the Ganges River Basin (including the Indian states of West Bengal, Bihar, Uttar Pradesh and Uttarakhand together with Nepal). However, during the course of the research, focus was also broadened to the Brahmaputra River Basin (BRB) in order to make comparisons between the two river basins in terms of arsenic groundwater concentrations. Focus was mainly put on the Indian part of the BRB, as data was only found for this part of the basin (including the Indian states of Sikkim, Assam, Arunachal Pradesh, Nagaland, Manipur, Manipur, Mizoram and Tripura). Therefore, this chapter will first introduce the GRB, followed by an overview of the BRM.

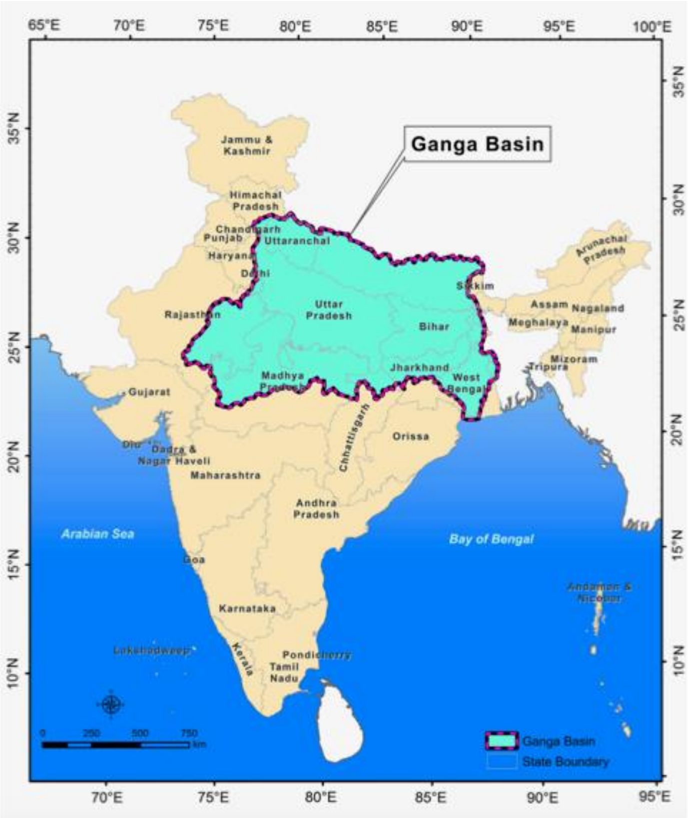

Figure 2: Location of the Ganges River Basin (Maheswaran et al., 2016).

Ganges River Basin

The GRB is a part of the Ganges-Brahmaputra-Meghna river basin, which houses a population of over 500 million people, draining a total of 1.08 million km^2 in the countries of China (Tibet), Nepal, India and Bangladesh whereby approximately 26% of India's land mass is covered. The GRB is considered as one of the most densely populated and fertile areas in the world and the basin's inhabitants rely directly or indirectly on the water resources of the GRB for food, drinking water and agriculture (Khan et al., 2012).

The origin of the Ganges River is the Bhagirathi which is found at an elevation of about 7000 m and the length is estimated at roughly 2520 km before the river eventually culminates into the Bay of Bengal. Several tributaries join the river on both sides of the riverbanks, throughout its traverse. The main tributaries are considered to be the Yamuna, the Ghaghra, the Gandak, the Kosi, the Mahananda and the Son (note that some of these have different names in Nepal). Snowmelt water originating from the Himalayas, return flow, base flow and precipitation-generated direct surface runoff account for the principal source of river water. The rainfall in the GRB is not uniform throughout its catchment. Besides changing throughout the region, the amount of rainfall received by the GRB is also largely limited to the few monsoon months of June until September/October. During the dry periods of November until May, low flow conditions can be observed in the Ganges River as well as its tributaries. Annual average rainfall rates differ from 350 mm in the western part to 2000 mm near the delta at the eastern part of the basin (Anand et al., 2018; Maheswaran et al., 2016).

The drainage area of the GRB is divided into many sub-basins (figure 4): (1) the Yamuna river which flows through the Indian states of Uttarakhand, Himachal Pradesh, Haryana, Uttar Pradesh and Delhi. The tributaries of the Yamuna river include the Betwa, Ken, Chambal, Tons and Sindh which partly drain the states of Madhya Pradesh and Rajasthan; (2) the Gomti river, flowing through Uttar Pradesh; (3) the Ghaghara river, which emerges from the Tibetan plateau and passes through Nepal, before crossing the states of Uttar Pradesh and Bihar; (4) the Gandak river, which flows through Nepal, Uttar Pradesh and Bihar; (5) the Kosi river, which passes through Nepal and Bihar; (6) The Sone river, crossing Madhya Pradesh, Uttar Pradesh, Jharkhand and Bihar; (7) the Punpun river, flowing through Jharkhand and Bihar; (8) the Damodar river, which passes through West Bengal and Jharkhand (Anand et al., 2018; Maheswaran et al., 2016).

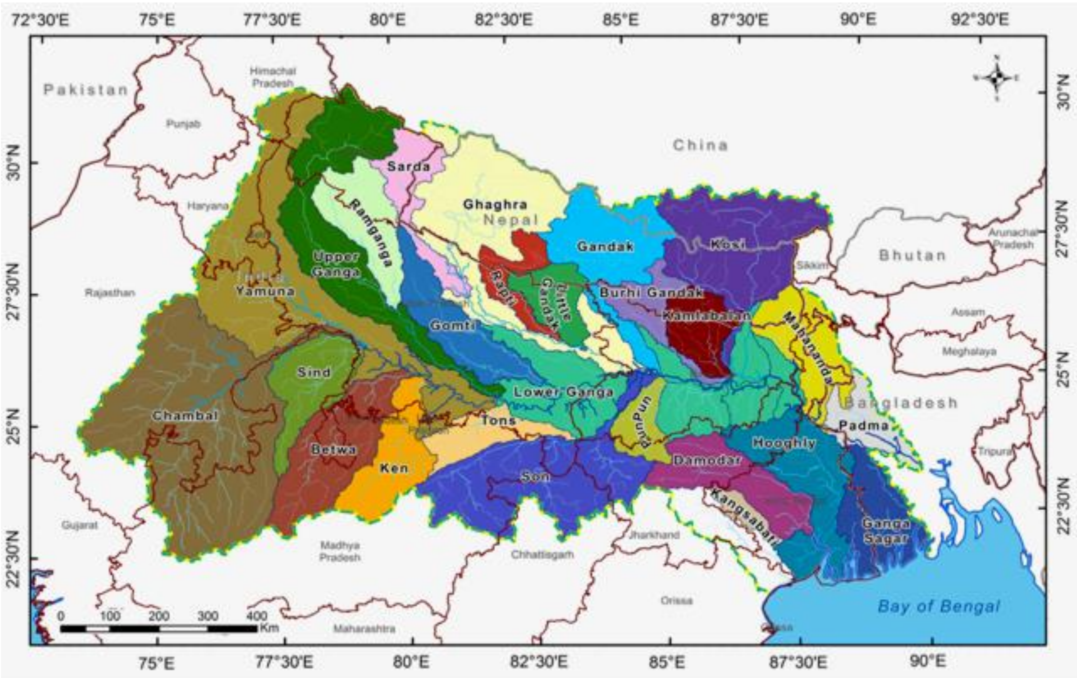

Figure 3: Sub-basins of the Ganges River drainage area (Maheswaran et al., 2016).

Geology of the GRB

The Himalayan foreland of the GRB is constructed of large stretched out floodplains, called the Gangetic Plains (figure 5), which act as active fluvial depositional basins. The width of these fluvial depositional basins stretches from roughly 200 km at the eastern part to about 450 km at the western part, whereas the length is estimated at approximately 1000 km in east-west direction (Singh, 1996).

The Gangetic Plains have been influenced by water regime, climate-driven sediment and intra- and extra-basinal tectonics (Sinha et al., 2005a). Compared to the degree of down-flexing of the basement, the sediment input in the Gangetic Plains has constantly been in an excess state (Singh, 2004a). Based on Garzanti et al. (2007) who stated that the upper Greater Himalayas (highest mountain range of the Himalayan Range (Searle et al., 2006)) and the Siwaliks devote up to 40% of the total sediment that is delivered to the Gangetic floodplains, Guillot et al. (2015) estimated that the Lesser Himalayas contributed a maximum of 45% whereas the Higher Himalayan Crystallines contributed only up to 15%.

Faults, depressions and a network of ridges criss-cross the basement of the Gangetic Plains. This basement is marked by three main subsurface ridges, being the Munger-Saharsa Ridge at the east, the Faizabad Ridge at the middle and the Delhi-Hardwar Ridge at the west (Sinha et al., 2005b). The Gangetic Plains can further be divided into the Upper Ganga Plain (UGP), the Middle Ganga Plain (MGP) and the Lower Ganga Plain (LGP) (figure 4), whereby the LGP merges with the deltaic plain of the GBM basin (Thomas et al., 2002). It is thought that the sedimentation of the LGP could have been affected by Pleistocene eustatic sea level-related base-level changes, letting the MGP and UGP remain without marine influences (Tandon et al., 2008).

The southern part of the MGP consists of Quaternary deposits, which are thickening in northerly direction and overlay the Precambrian basement. These deposits lie in the central and western parts of the MGP, whereas the Mio-Pliocene aged Gondwana and Rajmahal Traps (predominantly basalt rocks) are found between the Quaternary sediments and the Precambrian basement. The northern part of the MGP mainly consists of thicker Quaternary sediments (as compared to the southern Quaternary sediments) which adjoin the Siwalik Hills at the base of the Himalayas (Singh, 2004a). There is a striking resemblance regarding the geology of the MGP with that of the UGP, which has led to the beliefs that the arsenic problem affects the entire Gangetic Plain (Chakraborti et al., 2003).

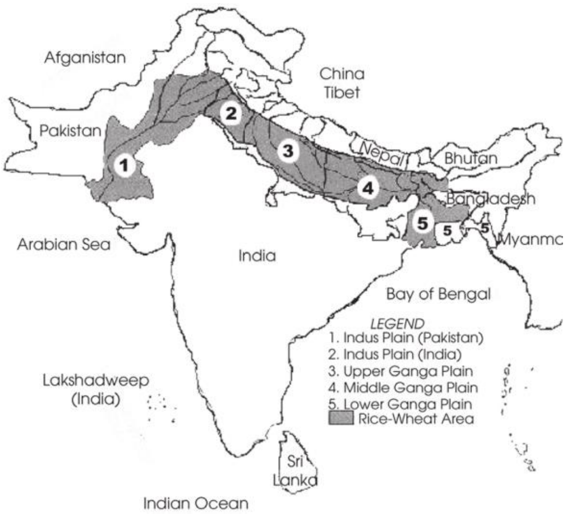

Figure 4: Location of the Indo-Gangetic Plains (Khurana et al., 2008).

Brahmaputra River Basin

The Brahmaputra is considered as a major transboundary river with a length of 3410 km, a drainage area of around 640,000 km^2 and an average discharge of approximately 21,000 m^3/s at the confluence with the Ganges in Bangladesh. The origin of the river is found in Southern Tibet (China) at the great glacier mass of Chema-Yung-Dung at an elevation of 5,300 m above sea level (m.a.s.l), located in the Kailas range. Here the river is named “Yarlung Tsangpo”, followed by “Siang” further downstream and eventually “Brahmaputra”. Before culminating into the Bay of Bengal, the Brahmaputra river traverses through the countries of China (1995 km), India (983 km), and Bangladesh (432 km) with tributaries also in Bhutan,. China accounts for 50.5% of the total catchment area, whereas India makes up 33.6%, Bangladesh is at 8.1% and Bhutan reaches up to 7.8% (Biswa et al., 2017). The BRB experiences diverse environments, including the dry and cold plateau of Tibet, the rain drenched slopes of the Himalaya, landlocked alluvial plains (Assam) and the vast Bengali deltaic lowlands (IWM, 2013; Gain & Wada, 2014).

The BRB (figure 6) can be categorized into three distinct physiographic zones: the Tibetan Plateau (TP), which covers 44.4% of the entire basin; the Himalayan belt (HB), which comprises 28.6%; and the floodplain (FP), which stretches over about 27%. China, India and Bhutan are part of the Upper BRB, which consists of the TP (elevation greater than 3500 m.a.s.l) and HB (elevation between 100 and 3500 m.a.s.l). The areas elevated lower than 100 m.a.s.l. are considered as part of the FP, which is seen as the Lower BRB and consists of parts of Bangladesh and India (Immerzeel, 2008). Similar to the GRB, the BRB has a monsoon driven climate, whereby the wet season, which lasts from June to September, produces 60-70% of the total yearly rainfall. The pre-monsoon season, which lasts from March to May, accounts for 20-25% of the total annual rainfall. The lower part of the basin has an average rainfall of 2354 mm/year (IWM, 2013).

The Brahmaputra river flows through one of the most densely populated areas of South Asia, where its water is used for agriculture, drinking water and energy by roughly 130 million people. In total, around 27500 million m^3 of water is used annually, of which India and Bangladesh use the most. The largest part of the river water is used for agriculture (89%), followed by domestic uses (9%) and industrial uses (2%) (IWM, 2013).

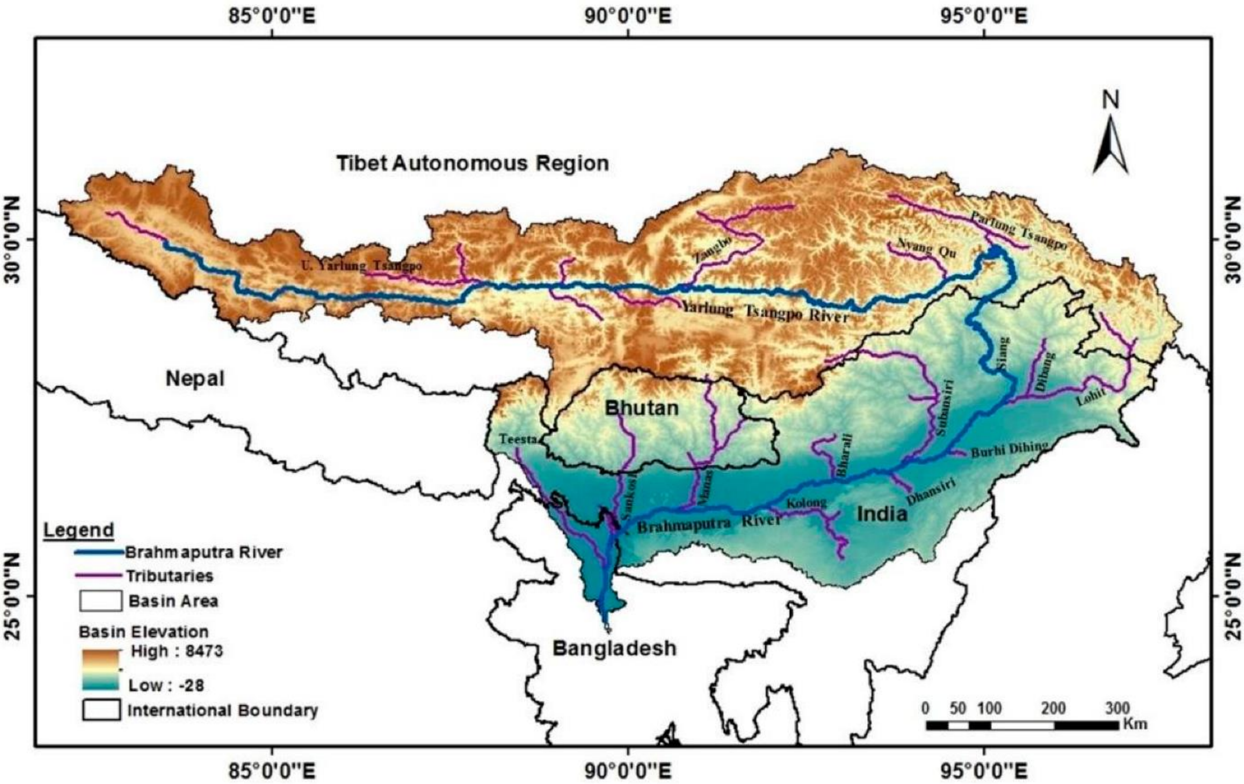

Figure 5: Topographic map of the Brahmaputra River Basin (Barua et al., 2019).

Geology of the BRB

The alluvial floodplain of the Brahmaputra in Assam is bordered by two different orogenic belts: the trans-Himalayan belt to the north and northeast and the Naga-thrust belt (Indo-Burmese range) in the south. These belts are continuing parts of the Indo-Gangetic-Brahmaputra foreland basin. The Eastern Himalayas surround the BRB on the east and north side, whereas the south is bounded by the Shillong Plateau and the Naga-thrust belt (Sing & France-Lanord, 2002).

In the Indus-Tsangpo suture zone, the Tsangpo and its tributaries (Doilung, Nyang Qu and Lhasa He), erode through volcanic and plutonic rocks of the Trans-Himalayan batholith (linked with Palaeozoic to Eocene age) (Goodbred et al., 2014). As the river reaches the eastern syntaxis (Namche Barwa) and flows into a southward direction, it enters a highly metamorphosed zone, associated with sediments and rocks of the Transhimalaya Plutonic Belt. This belt consists of two main units, being the Tidding Suture Zone and the Lohit Plutonic Complex (Singh & France-Lanord, 2002).

Tributaries of the Brahmaputra located in the east (such as the Dibang and Lohit) erode through these units (Singh et al., 2005). Many tributaries originate from the north and culminate into the Brahmaputra River in the floodplain of Assam. These tributaries include the Tipkai, Manas, Puthimari, Jia Bhareli, and Subansiri and they drain mostly metamorphic rocks and sedimentary sequences. The tributaries in the south (the Kopili, Burhi Dihing and Dhansiri) flow through the western half of the Naga-thrust belt, which is mostly composed of shales and sediments associated with ophiolites of Cretaceous and Oligocene age (Kumar, 1997).

Comparisons between the Ganga and Brahmaputra Rivers show that higher erosion rates occur in the eastern Himalayas (2.9 mm/year) than in the western Himalayas (2.1 mm/year). This is likely due to the eastern Himalayas having higher precipitation rates, which leads to a higher runoff in the BRB. Hereby, the erosion rates are directly controlled by the intensity of the monsoons (Galy & France-Lanord, 2001). Regarding the degree of weathering, a trend of more intense weathering over time has been observed in both the east and the west of the Himalayas. However, western Himalayan sediments appear to be generally more weathered compared to those in the east, despite the higher precipitation rates in the east. More extensive weathering of the western Himalayas is associated with a more seasonal climate which allows for physical weathering of sediments in the dry season. Meanwhile, more intense weathering in the eastern Himalayas is linked to the higher runoff rates which lead to increased rapid erosion and transport of sediments (Vögeli et al., 2017).

Elaboration on theories found in literature

Three main hypotheses exist on the primary source of arsenic in the Ganges and Indian Brahmaputra River Basins.

The Qamdo-Simao (QS) volcanic and ophiolite province is located north of the Namche Barwa syntaxis (figure 2) and can be described as “one of the gigantic metallogenic belts of the world” (Metcalfe, 1996). Stanger (2005) claimed that “if marine ferromanganoan-bound arsenic is to be concentrated anywhere in Asia, then the Permo-Triassic-Simao (or ‘Chamdo-Sze-Mao’ a.k.a. the ‘Sinjiang’ area (Xuanxue et al., 1994)) terrain suture/volcanic province (…) now constitutes the obvious likely reservoir”. Arsenic found in downstream reaches of the GBM basin is assumed to be derived indirectly from erosion of the Siwaliks or directly from erosion of the QS province. In both cases, ‘sedimentary dilution’ may locally decrease the sediment arsenic concentration (Stanger, 2005).

Guillot & Charlet (2007) disregarded the ideas of Stanger (2005) by claiming that the QS volcanic and ophiolite province cannot be the original source of arsenic in the Siwaliks and the adjacent laying

Himalayan floodplain, as the main rivers that originate from this province never flowed towards that direction. Therefore, there has to be another arsenic source and this potential area can be found in the Indus-Tsangpo suture zone (figure 2) which shares high similarities with the lithological characteristics of the QS province.

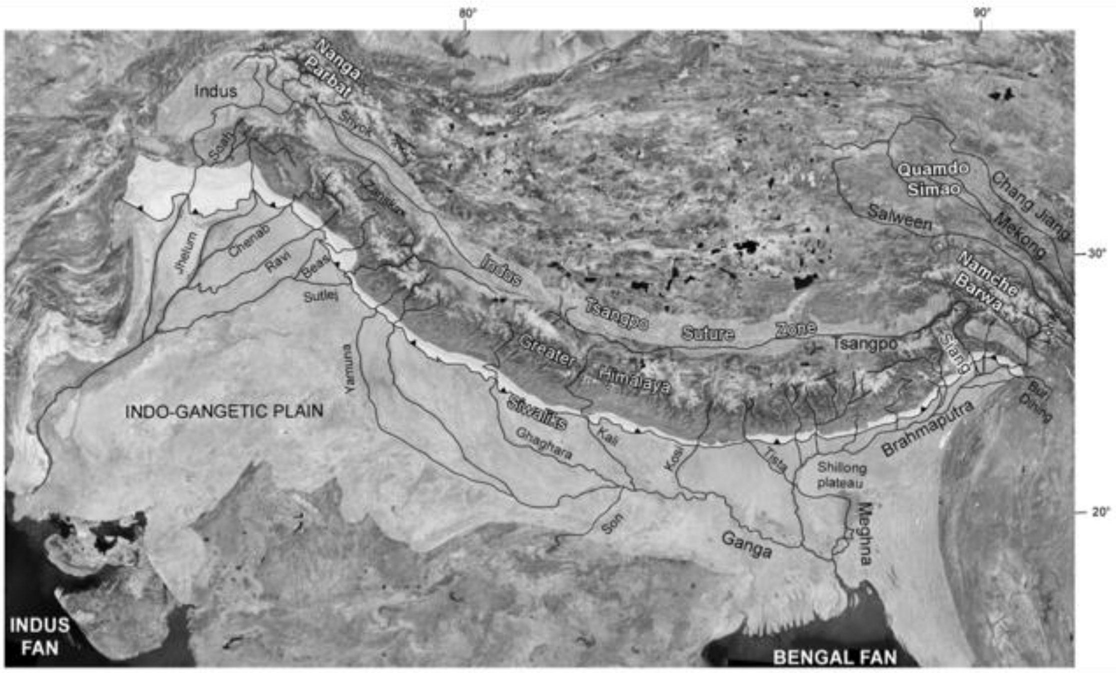

Figure 6: Himalayan orographic map showing the major Himalayan geologic zones and the main drainage systems (Guillot & Charlet, 2007).

Not many further theories exist on the potential primary source that supplied the Siwaliks with arsenic. However, Mueller (2018) claimed that the origin of arsenic in Nepal can be traced to a felsic initial source such as metapelites or leucogranites. The author stated that no ophiolites exist in the Nepalese Himalaya, therefore debunking the theories of Stanger (2005) and Guillot & Charlet (2007), which are based on the idea that ophiolites are the initial source of arsenic as contained in arsenopyrite. Mueller (2018) stated that the positive correlation of Na, Ka and trace elements such as Mo, B and Li with arsenic found in groundwater samples of the Terai, advocates against the widely approved hypothesis that the original source of arsenic is to be found in the mafic rocks which occur across the whole of the Himalayan belt. An original arsenic source in felsic rocks was reflected by observing typical felsic lithophile elements like U, Sr, P, B and Li. These elements are found ubiquitously in felsic rocks of the Nepalese Himalayas, such as metapelites and leucogranites which show a high abundance of As, P, B Cd and Pb (Mueller, 2018).

Furthermore, Guillot et al., 2015 carried out a geochemical and sedimentological study to investigate the origin of arsenic contamination in sediments in the Western Terai of Nepal, namely the Nawalparasi district. This study considered the correlation of major elements and rare earth elements found in sediments as deposited in the research area and its original source sediments. It was found that for the Late Pleistocene to Early Holocene sediments, the dominant source was the upper part of the Lesser Himalayan and the Higher Himalayan Crystallines, with the Siwaliks possibly having an input as well. Thus, it is assumed that the aquifer present in the Western Terai has a dominant arsenic source in the black schists of the Lesser Himalaya (Guillot et al., 2015).

3. Materials and methods

Methodology

The general setup of this research consisted of a data mining exercise whereby as many relevant literature sources as possible were used in order to construct a regional geographic image of the arsenic groundwater distribution in the research area. The first and second step of the methodology consisted of collecting (quantitative) data on concentrations of arsenic groundwater and heavy metals in the GRB and BRB (sub-questions 1 and 2). This was followed by the third step whereby the distribution of elevated arsenic groundwater was compared with the heavy metals distribution and linked to local surface geology (sub-question 3). The final step consisted of indicating possible arsenic sources by connecting features of the surface geology with the release of (groundwater) arsenic (sub-question 4).

Data collection and representation of results

The first step of this study consisted of collecting as many as possible elevated arsenic groundwater concentrations (anomalies) in the GRB, mainly focussing on the Indian states of West Bengal, Bihar, Uttar Pradesh, Uttarakhand and the country of Nepal. Additionally, arsenic groundwater data was also collected for the BRB in north-eastern India for the sake of comparison. In the interest of time, it was decided to not process any data of Bangladesh for this research. Although there has been extensive arsenic groundwater testing in Bangladesh (Kinniburgh & Smedley, 2001), leaving this area out was mainly done as the floodplains of Bangladesh act as a major sink of arsenic. Hence, a primary source of arsenic should rather be found further upstream in the two basins.

The search engine Google Scholar was mainly used to search for relevant reports and studies related to arsenic groundwater concentrations, with using search terms such as "arsenic groundwater GRB", "arsenic groundwater BRB", "arsenic groundwater Nepal" etc. However, also the normal google search engine was used to find "grey data" and reports published by institutes which were focused on arsenic groundwater testing. For example, the Central Ground Water Board (CGWB) provided useful data for India. In order to obtain more detailed data of arsenic groundwater concentrations in the Nepal Himalaya, various institutes located in Nepal (which conducted arsenic groundwater testing in this country), were contacted. These contacted institutes included: UNICEF, WHO, Nepal Red Cross Society (NRCS), Nepal Water Supply Corporation (NWSC), Nepal Water for Health, Rural Water Supply and Sanitation Fund Development Board (RWSSFDB), Department of Water Resources and Irrigation (DWRI), Japan International Cooperation Agency (JICA) and Public Health Organization (ENPHO). Unfortunately, contacting these institutes turned out to be ineffective, as none of them wished to share detailed data.

To visualize the distribution of the arsenic groundwater concentrations, the data was presented in a geospatial map, which was constructed using ArcGIS Pro. The reports and associated data that were used for constructing this map, are shown in table 1. The number of samples taken differed significantly per each report, ranging from 4 to over 1,000,000 samples. Since the different reports differed significantly in amount of samples per report, a statistical analysis of the data was not carried out. For the sake of structure and visualisation, not every sample location, as found in the literature, were used as a single data point in the geospatial map. Sample locations of a single study were either represented as individual data points or aggregated to data points showing maxima values (table 1). Hence, not every data point as visualized in the geospatial map is based on the same amount of arsenic groundwater samples and should rather be seen as indicators instead of qualitative data points. The maxima of arsenic groundwater concentrations were classified into three categories: low (< 10 μg/L), intermediate (10 – 50 µg/L) and high (> 50 µg/L), based on the WHO (10 µg/L) and Indian and Nepal (50 µg/L) guidelines. The first step also aimed to collect data of areas that indicated no elevated groundwater concentrations. In the geospatial map, the sample locations which indicated no elevated arsenic groundwater concentrations were categorized in a range of 0 – 10 µg/L. In the case of the situation where the sample locations were not shown or explained in the literature, a polygon was constructed to broadly represent the studied area.

Table 1: Reports and associated data used for visualisation.

Report	Number of samples	Data points used
Singh et al., 2018	136	8 (aggregated data)
Mehrotra et al., 2014	150000	5 (aggregated data)
Rahman et al., 2021	93	22 (aggregated data)
Saxena et al., 2014	13	1 (aggregated data)
CGWB, 2018	510	376 (individual observations)
Shrestha et al., 2014	61	6 (aggregated data)
Emerman et al., 2013	52	52 (individual observations)
Emerman et al., 2014	24	24 (individual observations)
Ghezzi et al., 2017	10	10 (individual observations)
Kumar et al., 2019	35	9 (aggregated data)
Sharma et al., 2016	4	1 (aggregated data)
Brikowski et al., 2016 (and references here in)	>1000000	130 (aggregate data)
Singh, 2004b; Singh, 2007	848	64 (aggregated data)
Ghezzi et al., 2019	9	9 (individual observations)
Chhimwal et al., 2022; Gupta et al., 2012	39	13 (aggregated data of averages, represented in polygon)
Dongol et al., 2005	38	1 (aggregated data)
Yadav et al., 2015	Not specifically mentioned	1 (aggregated data)
Shrestha et al., 2017	Not specifically mentioned	2 (aggregated data)
Aryal et al., 2012	84	1 (aggregated data)
Bhusal & Gyawali, 2015	30	12 (aggregated data)

The second step was carried out simultaneously with the first step. This step consisted of finding data concerning concentrations of heavy metals which most commonly occur in sulphides like Fe, Cu, Pb, Co, Zn and Ag. It was aimed to collect data on concentrations of the heavy metals in the same regions as the arsenic groundwater concentrations. In order to do so, data was retrieved from the Indian Institute of Technology (ISM), Uttarakhand Public Service Commission (UKPSC), Bureau of Land Management (BLM), Indian Bureau of Mines (IBM) and Nepal Geological Society (NGS). Instead of actual concentrations of Fe, Cu, Pb, Co, Zn and Ag, the data provided locations where these heavy metals are mined from ore minerals, therefore rather giving an indication of anomalies of the concerned heavy metals. Similar to the first step, the data was presented in a geospatial map constructed in ArcGIS Pro, whereby the locations which indicated heavy metal anomalies, were represented as data points.

Interpretation of results

For the third step, the arsenic anomalies in the GRB and Indian BRB were first compared to the distribution of the heavy metals, whereby it was aimed to find out whether or not there was a correlation between both sets of data. Next, the arsenic groundwater concentrations were linked to the local surface geology by using the following geological maps (Appendix B1 to B10):

- Geological map of West Bengal (Bandyopadhay et al., 2014)
- Geological map of Bihar (Roy, 2017)
- Geological map of Uttar Pradesh (MoEF, 2011)
- Geological map of southern Uttar Pradesh (Dinkar et al., 2019)
- Geological map of Uttarakhand (Das & Modak, 2019)
- Geological map of Nepal (HMH, n.d.)
- Geological map of Northeast India (Verma et al., 2016)
- Geological map of Sikkim (Baruah et al., 2019)
- Geological map of Mizoram (Bharali et al., 2017)
- Geological map of the Marsyangdi river basin (Ghezzi et al., 2019)
- Geological map of Jhikhu Khola catchment (Dongol et al., 2005)

For the final step, possible primary arsenic sources in the GRB were indicated as based on features (such as mineralogy) of the surface geology. This potential arsenic source was then further discussed in terms of supporting evidence (indication by trace elements) and it was argued whether this source could also contribute to the arsenic anomalies in the Indian BRB.

4. Results

This chapter first describes the arsenic groundwater anomalies found in the GRB and BRB, followed by an description of the areas with only low arsenic groundwater concentrations and lastly, a description of the heavy metal anomalies.

Arsenic groundwater anomalies in the GRB

The arsenic maxima as classified in categories of intermediate (10 – 50 µg/L), and high (> 50 µg/L) are shown in figure 7. Arsenic anomalies of were found in the Indian states of West Bengal, Bihar, Uttar Pradesh and Uttarakhand. Additionally, arsenic anomalies were also found in various catchment areas in Nepal. This section will first describe the arsenic groundwater anomalies found per Indian state, followed by a description of the arsenic anomalies in the different Nepali catchments. Extended data on the maxima of arsenic groundwater concentrations for specific districts in West Bengal, Bihar, Uttar Pradesh and Uttarakhand are shown in Appendix C1 to C4.

West Bengal

In the state of West Bengal, arsenic groundwater concentrations per district were found with maxima ranging up to 405 µg/L (table 2; Appendix C1). Maxima in the intermediate category were found in 6 districts (Hooghly, Malda, Murshidabad, Nadia, North 24 Parganas and South 24 Parganas), whereas only 3 districts (Burdwan, Howrah and Kochbihar) contained maxima of the high category.

Bihar

Concerning the arsenic groundwater concentrations per district in the state of Bihar, 19 districts were found to have maxima in the intermediate category. Only the districts of Dhanbad and Godda were found to have maxima of the high category, with the maximum values in this districts being 57.0 µg/L and 60.0 µg/L, respectively (table 2; Appendix C2).

Uttar Pradesh

The maxima of arsenic groundwater concentrations per district in Uttar Pradesh were found to be intermediate in 7 districts, whereas high maxima were found in the districts of Azamgarh, Bahraich, Deoria, Lakhimpur and Maunath Bhanjanm, with the district of Azamgarh having the highest maximum value of 811 µg/L (table 2; Appendix C3).

Uttarakhand

In the state of Uttarakhand, only the district of Haridwar contained elevated (i.e. > 10 µg/L) maxima of arsenic groundwater concentrations. In the Haridwar district, arsenic groundwater concentrations were found to have a maximum value of 84.0 µg/L in the Laksar block and a mean value of 12.5 µg/L in the Bhagwanpur block (table 2; Appendix C4).

Table 2: Ranges in elevated (> 10 µg/L) maxima of arsenic groundwater concentrations in the Indian states of West Bengal, Bihar, Uttar Pradesh and Uttarakhand (CGWB, 2018; Sharma et al., 2016; Kumar et al., 2019; Singh et al., 2018; Mehrotra et al., 2014; Rahman et al., 2021 Saxena et al., 2014).

State	**As (µg/L)**
West Bengal	10.0 – 405
Bihar	10.0 – 57.0
Uttar Pradesh	10.0 – 811
Uttarakhand	10.0 – 84.0

Nepal

Figure 7 shows that elevated maxima of arsenic groundwater concentrations were found in almost all districts of the Terai, only the district of Jhapa in the far east of Nepal was found clear of intermediate and high maxima. Furthermore, only the district of Morang was found to only have intermediate maxima. The most affected regions appear to be Nawalparasi, Kanchanpur, Kailali, Bardya, Parsa and Bara.

In central Nepal, arsenic groundwater anomalies were found in Pokhara Valley and Mustang Valley (figure 8). Arsenic groundwater concentrations in Pokhara Valley were found to generally range from 8 – 810 µg/L, with some outliers ranging up to 1220 – 7900 µg/L. Mustang Valley was found to have arsenic groundwater concentrations between 0 – 436 µg/L. In eastern Nepal (figure 9), arsenic groundwater anomalies were found in Kathmandu Valley and the towns of Charikot (Dolkha district) and Manthali (Ramechhap district). In Kathmandu Valley, elevated maxima of arsenic groundwater concentrations were found to range from 10 to above 50 µg/L. A range of 10 – 50 µg/L in arsenic groundwater concentrations was found in the towns of Charikot and Manthali. Lastly, arsenic groundwater concentrations were found to range from 3.0 – 48.0 µg/L in the Arthunge Municipality in the district of Myagdi (table 3).

Table 3: Ranges in arsenic groundwater concentrations in Nepal: Terai and Kathmandu Valley show ranges of elevated maxima, whereas Pokhara Valley, Mustang Valley, Charikot & Manthali and Arthunge Municipality show actual ranges of individual samples (Shrestha et al., 2014; Emerman et al., 2013; Emerman et al., 2014; Shrestha et al., 2014; Aryal et al., 2012; Ghezzi et al., 2017).

Location	**District**	**As (µg/L)**
Terai	Kanchanpur, Kailali, Bardiya, Banke, Dang, Kapilvastu, Rapandehi, Nawalparasi, Chitwan, Parsa, Bara, Rautahat, Sarlahi, Mahottari, Dhanusa, Siraha, Saptari, Sunsari and Morang	10 – > 50 µg/L
Kathmandu Valley	Kathmandu, Lalitpur and Bhaktapur	10 – > 50 µg/L
Pokhara Valley	Kaski	8 – 810 µg/L (outliers: 1220 – 7900 µg/L)
Mustang Valley	Mustang	0 – 436 µg/L
Charikot and Manthali	Dolkha and Ramechap	10 – 50 µg/L
Arthunge Municipality	Myagdi	3.0 – 48.0 µg/L

Arsenic groundwater anomalies in the BRB (Northeastern India)

In the BRB, arsenic groundwater anomalies were solely found in (Northeastern) India, as no data on arsenic groundwater concentrations were found in Tibet and Bhutan. The affected Indian states include Assam, Manipur, Arunachal Pradesh, Tripura and Nagaland, as depicted in figure 10. Extended data on the arsenic groundwater concentrations for specific districts in the Northeastern Indian states are presented in Appendix C5.

For the state of Assam, elevated maxima of arsenic groundwater concentrations were found in 12 districts and range from 10.0 – 657 µg/L, with only two districts (Sibsagar and Sonitpur) having intermediate maxima. The only district in the state of Manipur with elevated maxima of arsenic groundwater concentrations is the Thoubal district with a range of 798 - 986 µg/L. Six districts in the state of Arunachal Pradesh were found to have elevated maxima of arsenic groundwater concentrations, where all of these districts had high maxima with an overall range of 58 – 618 µg/L. Elevated maxima of arsenic groundwater concentrations in the state of Tripura were found in three districts with an overall range of 191 – 444 µg/L. Only two districts in the state of Nagaland were found to have elevated maxima of arsenic groundwater concentrations, ranging from 159 – 278 µg/L (table 4).

Table 4: Ranges in maxima of arsenic groundwater concentrations for states in Northeastern India (CGWB, 2018; Singh, 2004b; Singh 2007).

State	**As (µg/L)**
Assam	10.0 – 657
Manipur	798 – 986
Arunachal Pradesh	58.0 – 618
Tripura	191 – 444
Nagaland	159 – 278

Areas having arsenic groundwater concentrations only < 10 µg/L

Areas containing only arsenic groundwater concentrations below 10 µg/L are shown in figures 7 and 10. These areas are located in the Indian states of Uttarakhand, Sikkim and Mizoram, as well as various catchment areas in Nepal.

In the state of Uttarakhand, it was found that all 13 districts had groundwater containing arsenic below 10 µg/L, with a range of 0.17 – 2.50 µg/L (table 5). The states of Sikkim and Mizoram had no specific measurement locations indicated in the literature (Singh, 2007), whereby the state of Sikkim was listed as having arsenic groundwater concentrations of < 2.0 µg/L and Mizoram having arsenic groundwater concentrations of < 10.0 µg/L. See appendix C6 for extended data that is summarised in table 5

Table 5: Ranges in averages of arsenic groundwater concentrations for Indian states containing only arsenic below 10 µg/L (Gupta et al., 2012; Chhimwal et al., 2022; Singh, 2007).

State	**As (µg/L)**
Uttarakhand	0.17 – 2.50
Sikkim	< 2.0
Mizoram	< 10.0

In Central Nepal, areas with arsenic groundwater concentrations only below 10 µg/L were found in the Badigad catchment located in the Gulmi and Baglung districts as well as the Marsyangdi catchment in the Manang and Lamjung districts. The actual range in low arsenic groundwater concentrations in the Badigad catchment was not presented in the literature as all groundwater samples were listed as having a arsenic concentration of < 10.0 µg/L (Bhusal & Gyawali, 2015). In the Marsyangdi catchment, low arsenic groundwater concentrations were found to generally range from 0.18 – 3.34 µg/L. In eastern Nepal, low arsenic groundwater concentrations were found in the Dhankuta Municipality of Dhankuta district and in the Jhikhu Khola catchment in the Kavrepalanchok district. Also in the Dhankuta Municipality, the literature did not provide the actual range in low arsenic groundwater concentrations; all groundwater samples taken here, were listed as having a arsenic concentration of < 5.0 µg/L (Dongol et al., 2005). No groundwater samples containing arsenic were found in the Jhikhu Khola catchment (figures 8 and 9; table 6).

Table 6: Nepalese locations with ranges in arsenic groundwater concentrations only below 10 µg/L (Bhusal & Gyawali, 2015; Yadav et al., 2015; Ghezzi et al., 2019; Dongol et al., 2005).

Location	**District**	**As (µg/L)**
Badigad catchment	Gulmi and Baglung	< 10.0
Dhankuta Municipality (Koshi catchment)	Dhankuta	< 5.0
Marsyangdi catchment	Manang and Lamjung	0.18 – 2.92 (cold waters) < 1.0 – 3.34 (hot waters) (single outlier of 37.8)
Jhikhu Khola catchment	Kavrepalanchok	0.0

Heavy metal anomalies

The distribution of heavy metal anomalies in the GRB is depicted in figure 11. In India, anomalies of the heavy metals are distributed over the states of West Bengal, Bihar and Uttarakhand. West Bengal was found to have anomalies of iron, copper, silver and lead + zinc. The state of Bihar was also found to have anomalies of iron, copper, silver and lead + zinc. In Uttarakhand, it was found that anomalies of iron, lead and silver occur in this state (IBM, 2010; BLM, 2022; UKPSC, 2022; ISM, 2017). In Nepal, anomalies were found of iron, copper, zinc + lead, cobalt, nickel and silver (Kaphle, 2020).

The distribution of heavy metals in the BRB in Northeastern India is presented in figure 12. It was found that the state of Nagaland has an anomaly of nickel. Additionally, copper anomalies occur in the state of Manipur as well as in the state of Arunachal Pradesh (BLM, 2022).

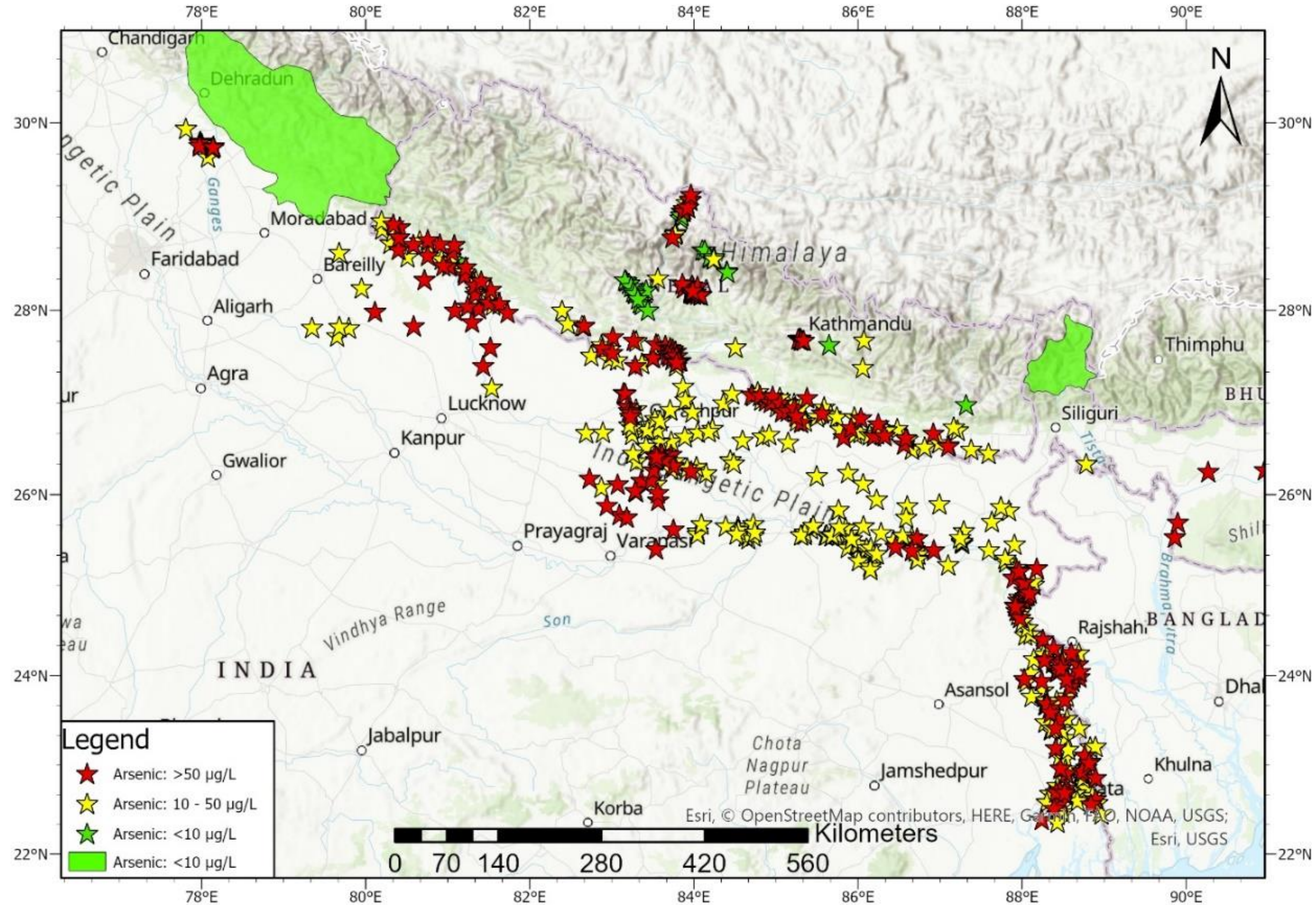

Figure 7: Maxima of arsenic groundwater concentrations in the Ganges River Basin, as categorized into low (< 10 µg/L), intermediate (10 – 50 µg/L) and high (> 50 µg/L). Note that data of the Terai region is aggregated of a single study project.

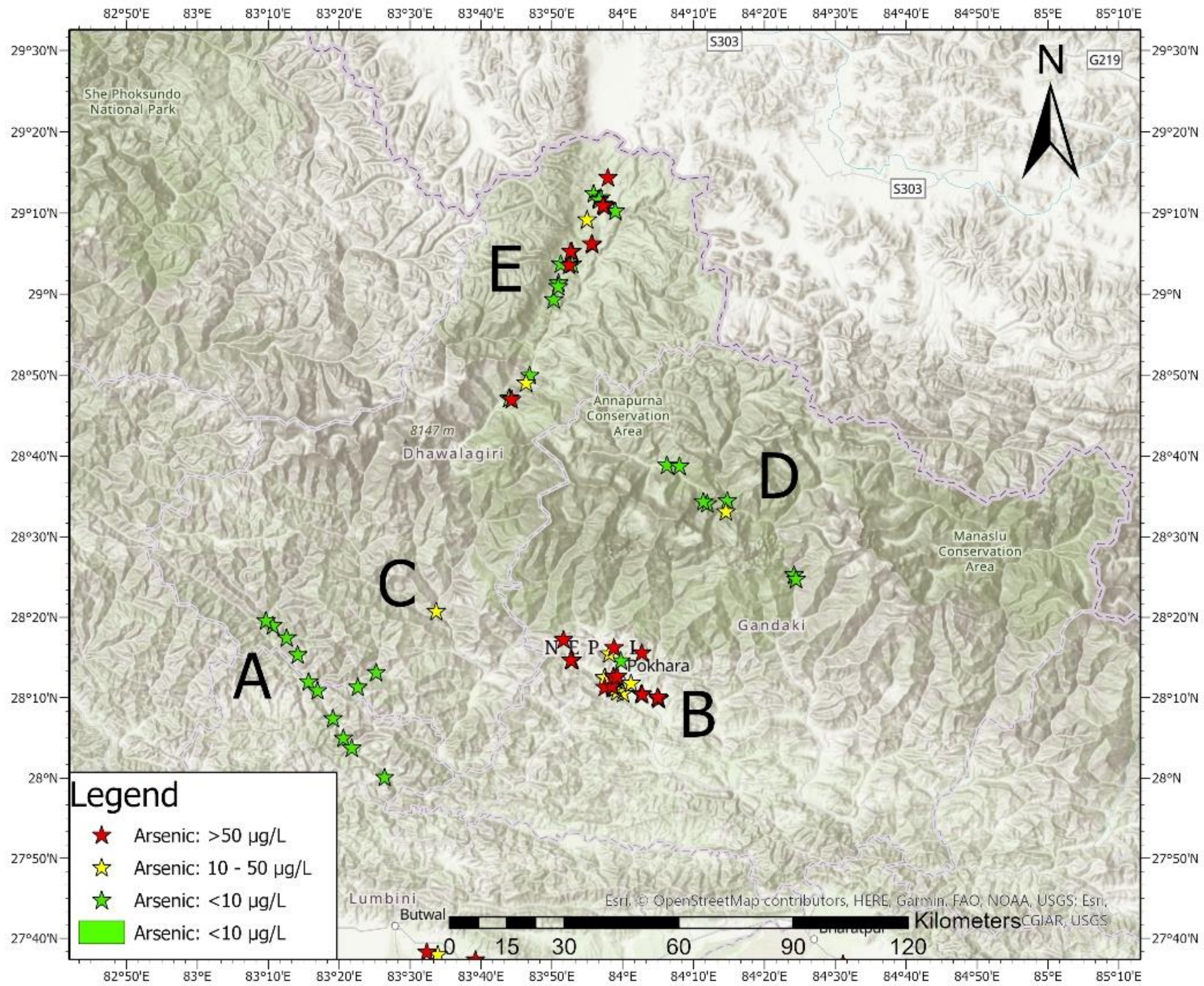

Figure 8: Individual observations of arsenic groundwater concentrations in central Nepal, as categorized into low (< 10 µg/L), intermediate (10 – 50 µg/L) and high (> 50 µg/L). A: Badigad catchment; B: Pokhara Valley; C: Arthunge Municipality; D: Marsyangdi catch

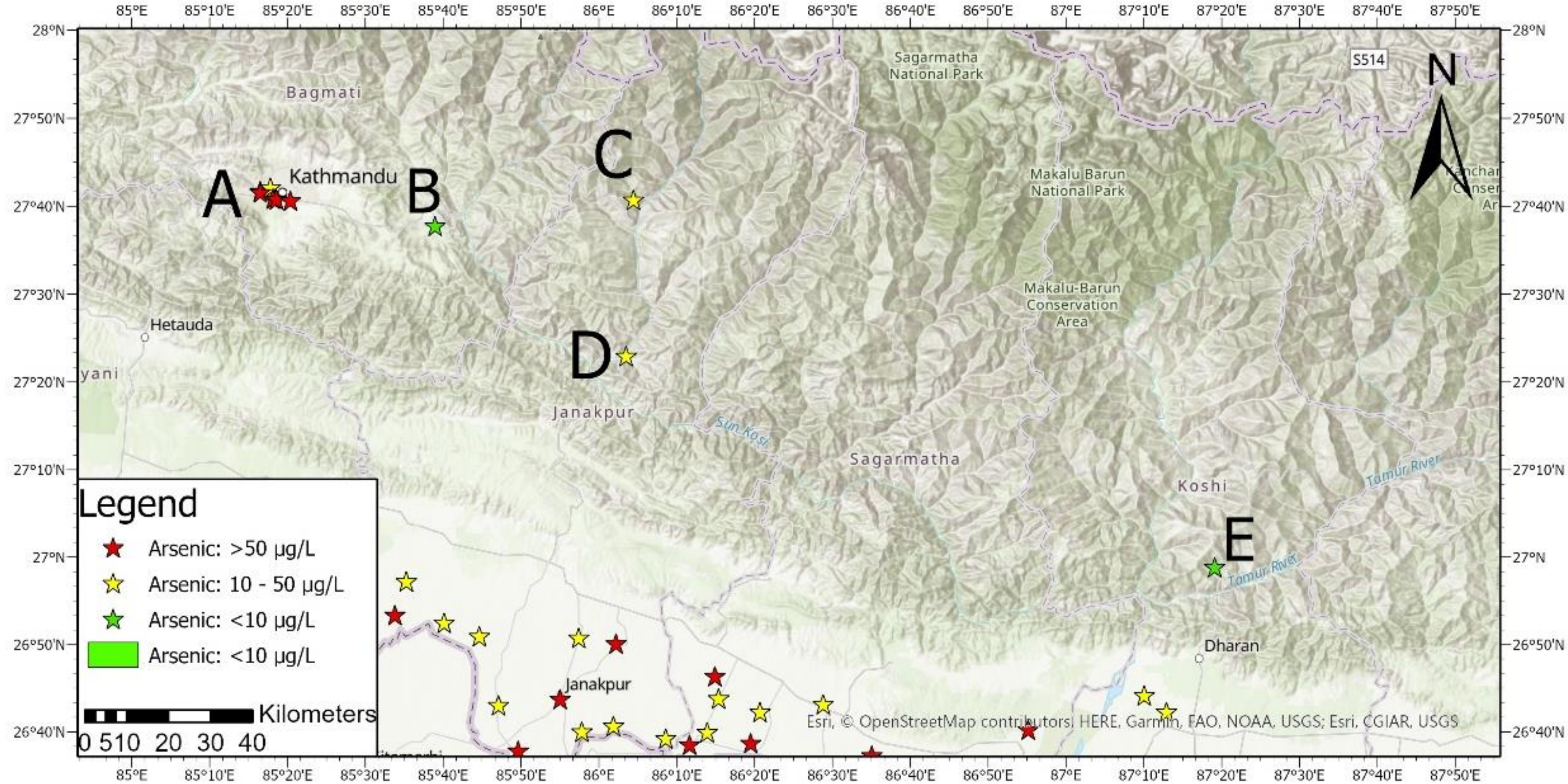

Figure 9: Maxima of arsenic groundwater concentrations in eastern Nepal, as categorized into low (< 10 µg/L), intermediate (10 – 50 µg/L) and high (> 50 µg/L). A: Kathmandu Valley; B: Jhikhu Khola catchment; C: Charikot; D: Manthali; E: Dhankuta Municipality. Note that data for B, C, D and E are maxima values of aggregated data.

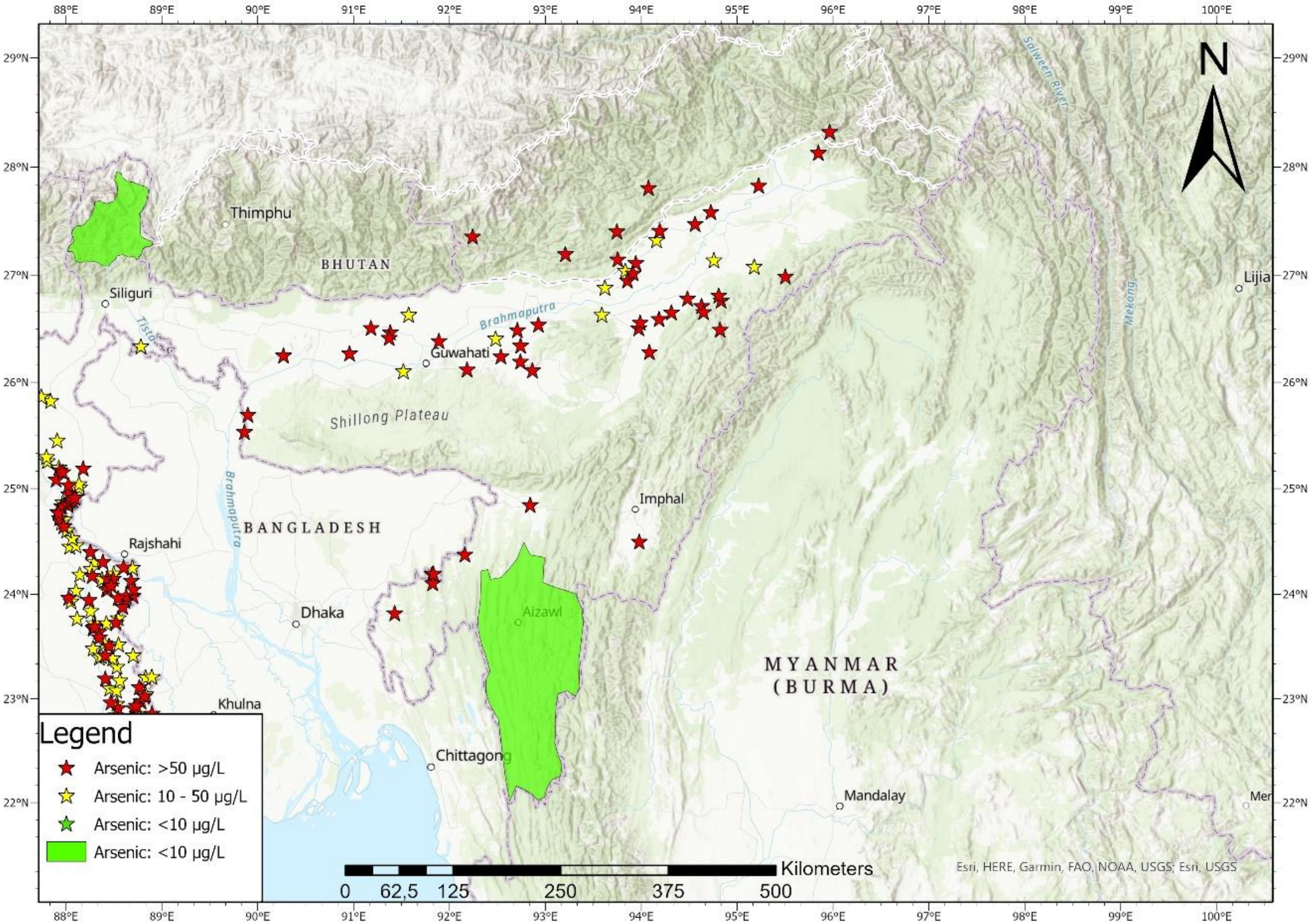

Figure 10: Maxima of arsenic groundwater concentrations in the Brahmaputra River Basin, as categorized into low (< 10 µg/L), intermediate (10 – 50 µg/L) and high (> 50 µg/L).

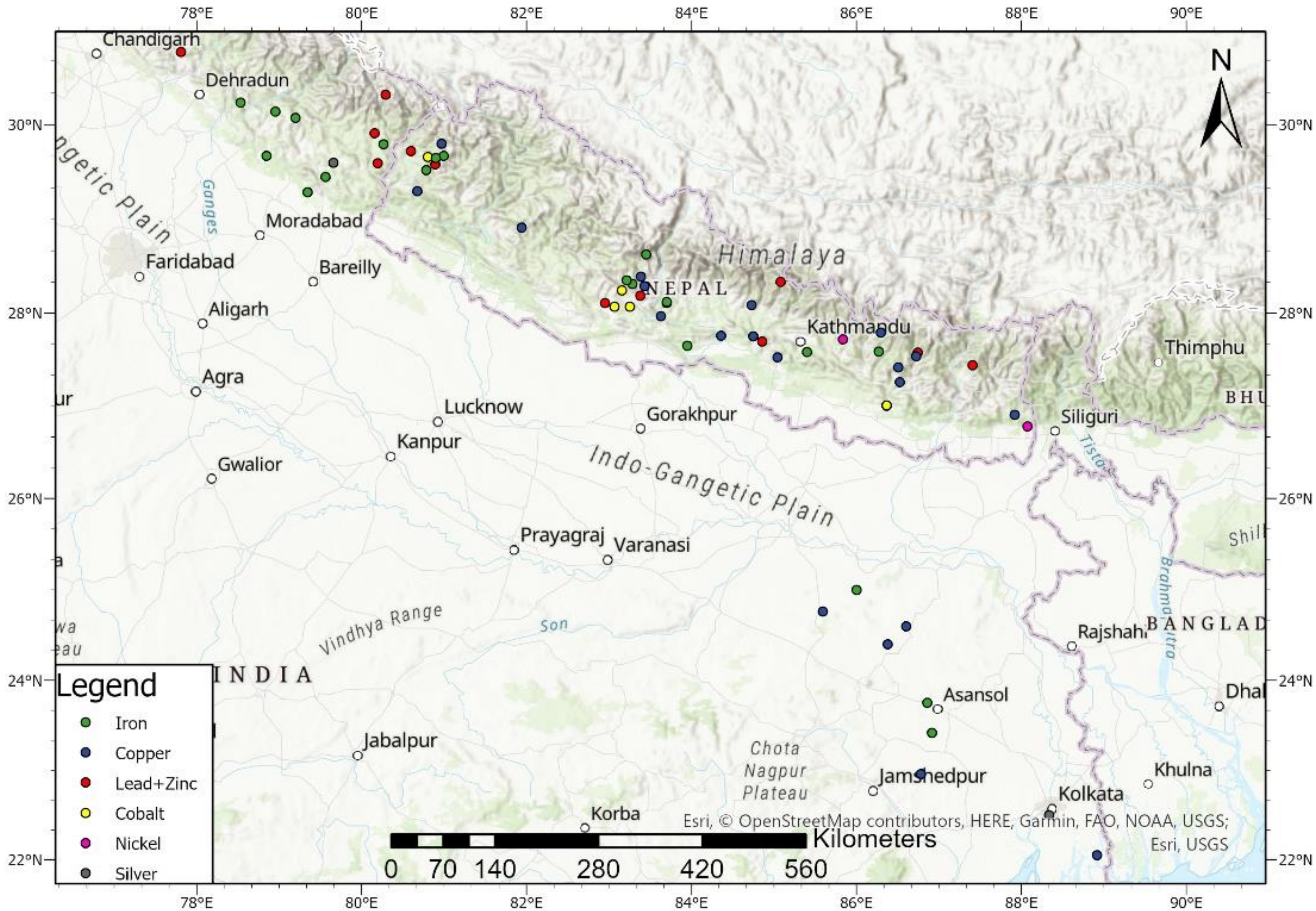

Figure 11: Distribution of heavy metal anomalies in the Ganges River Basin.

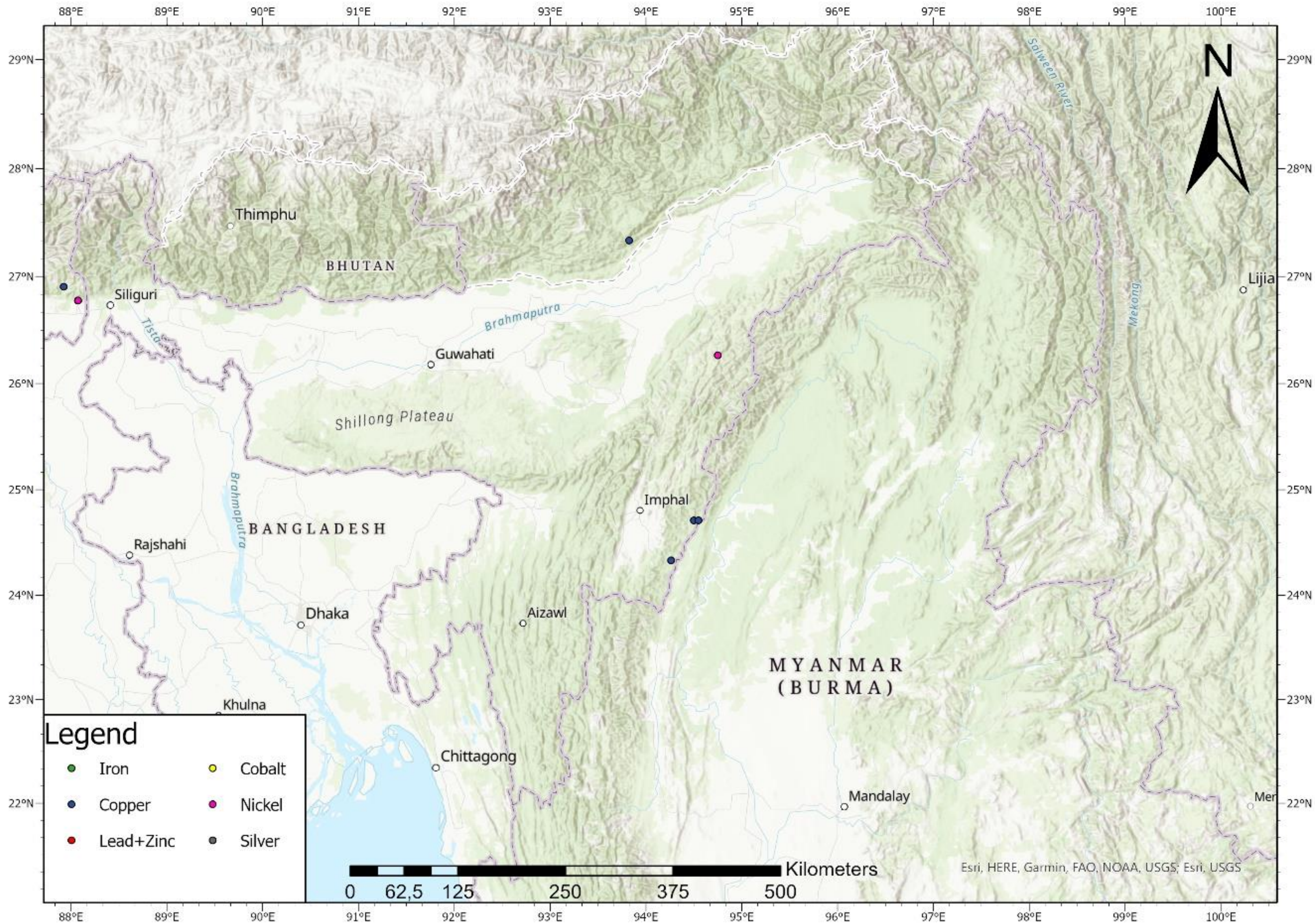

Figure 12: Distribution of heavy metal anomalies in the Brahmaputra River Basin.

5. Discussion

This chapter discusses the following points: the correlation of heavy metal anomalies with arsenic groundwater anomalies; the differences in surface geology between areas containing arsenic anomalies and areas having low arsenic groundwater concentrations; indication of primary arsenic sources; testing of the hypotheses; recommendations for further research; and limitations of the current study.

Correlation of heavy metal anomalies with arsenic groundwater anomalies

When comparing the heavy metal anomalies distribution maps with the maps showing the arsenic groundwater anomalies, there does not seem to be a clear correlation between the occurrence of heavy metal anomalies and the presence of elevated groundwater arsenic concentrations in the research area. This implies that the occurrence of heavy metal anomalies associated with sulphides is not a clear indicator for elevated arsenic groundwater concentrations. Hence, the occurrence of heavy metal anomalies cannot be used as proxy for the source of arsenic. Various studies carried out in Kathmandu Valley (Emerman et al., 2010), Pokhara Valley (Emerman et al., 2013) and Mustang Valley (Emerman et al., 2014) suggested that their geochemical data were inconsistent with the sulphide-oxidation model (Smedley, 1996), which is in line with the weak correlation between sulphide-associated heavy metal anomalies and arsenic groundwater anomalies, as found in this study.

Geological links

This section aims to identify whether there is a difference in local surface geology between areas with arsenic anomalies and areas with only low arsenic groundwater concentrations.

Areas having arsenic anomalies (in the GRB and Indian BRB)

Extended data on the surface geology of arsenic affected districts in West Bengal, Bihar, Uttar Pradesh and Northeastern India is presented in appendix D1 to D4 for background information.

West Bengal

The state of West Bengal is located in the Lower Ganga Plain and hence part of the GRB floodplain of (Singh, 2004a). The geological environment does not variate much in this state as most of the arsenic affected districts are associated with the Newer alluvium and Older alluvium formations (table 7). The sediments of these alluvia are of Holocene and Middle to Upper Pleistocene age, respectively. The Holocene Newer alluvium deposits consist mostly of silt, clay, sand and peat deposits, whereas the Pleistocene Older alluvium deposits contain mainly silt, sand and clay sediments (Bandyopadhay et al., 2014).

Bihar

For the state of Bihar, the geological setting is even less varying compared to the geology of West Bengal, as only the district of West Champaran (located in the southwest of Bihar) contains partly Older alluvium deposits (Quaternary to Upper Tertiary) as well as sediments of the Vindhyan Supergroup (of Lower Cambrian to Proterozoic age). This Vindhyan Supergroup is largely formed of sandstone, limestone and dolomite. The rest of the indicated arsenic affected districts contain Newer alluvium sediments (table 7). The alluvia in Bihar have similar sediments compositions as the ones in the state of West Bengal, as the Newer alluvium in Bihar consists of gravel, sand, clay, silt, calcareous concretions and pebble, whereas the Older alluvium consists of mostly sand, silt, ferruginous concretions, clay, pebble, cobbles and gravel (Roy, 2017).

Uttar Pradesh

Similar to West Bengal and Bihar, most of the indicated arsenic groundwater affected districts in the state of Uttar Pradesh are part of the Newer alluvium (table 7). In Uttar Pradesh, the Newer alluvium consists of mainly (locally micaceous) sand, pebble, silt and clay. The southwestern district of Jhansi is the only district that contains the Bundelkhand Granitoid Complex of Archean to Mesozoic age. This formation has a lithology of coarse to fine grained porphyritic granite and fine to medium grained leucogranite (MoEF, 2011; Dinkar et al., 2019).

Uttarakhand

The indicated arsenic groundwater affected locations, Bhagwanpur and Laksar blocks within Uttarakhand, are both located in the Newer and Older alluvium (Table 7). The sediment composition of these alluvia in this state does not differ from the other Indian states in the GRB and is mainly composed of sand, silt, clay and gravel (Das & Modak, 2019; Dinkar et al., 2019).

Table 7: Surface geology of the studied arsenic affected Indian states in the GRB.

State	**Surface geology**	**Sediments and rock characteristics**
West Bengal	Newer alluvium + Older alluvium	Silt, clay, sand and peat
Bihar	Newer alluvium + Older alluvium, Vindhyan Supergroup	Sand, silt, clay, pebble, cobbles, pebble and gravel. Sandstone, limestone and dolomite
Uttar Pradesh	Newer Alluvium, Bundelkhand Granitoid Complex	Sand (locally micaceous), silt and clay. Porphyritic granite and leucogranite
Uttarakhand (Bhagwanpur and Laksar block)	Newer alluvium + Older alluvium	Sand (locally micaceous), silt, clay and gravel

Nepal

The surface geology of the Terai region has a similar lithology as the Newer and Older alluvia of the Gangetic Plains. A rock type of gravels, sands and clays belongs to an alluvial surface geology (of Quaternary age), whereas conglomerates, sandstones, clays and shales form the bulk lithology of the Upper, Middle and Lower Siwalik Formations (Mid Miocene to Pleistocene age) (HMH, n.d.).

The geological setting of the Kathmandu Valley is mainly made up of the Chandragiri and Tistung Formations, both belonging to the Kathmandu Group (Pre-Cambrian to Devonian age). According to HMH (n.d.), the Chandragiri Formation consists mostly of fine-grained crystalline limestones with quartzites in the upper parts, whereas the Tistung Formation is mainly composed of phyllites, sandstones and sandy limestones (table 8).

Pokhara Valley contains mainly the Seti Formation and the Ranimatta Formation, associated with the Pokhara Subgroup and Dailekh Subgroup (both of Upper Pre-Cambrian to Late Palaeozoic age). These two formations have a highly similar lithology, as the Seti Formation consists of chlorite and muscovite sandstones, gritstones with conglomerates, massive quartzites in the upper parts and noted basic intrusions. Meanwhile, the Ranimatta Formation is composed of gritty phyllites, gritstones with conglomerates and massive quartzites in the upper parts where basic intrusions are abundant (HMH, n.d.).

In Mustang Valley, the surface geology is composed of the Higher Himalayan Complex and Tibetan Sedimentary Zone. The Higher Himalayan Complex has a sedimentology of mainly fluvial and fluvio torrential sediments with local lacustrine clays and marlstones (Pliocene to Holocene age), whereas the Tibetan Sedimentary Zone (Triassic to Lower Cretaceous) is characterized by shallow continental platform sediments with locally occurring pro-delta facies. Slates, sandstones and shales with glauconite are also present (HMH, n.d.). A formation which is part of the Higher Himalayan Complex surface geology is the Thakkhola Formation (Dhital, 2015). This formation is composed of fluvial and alluvial fan conglomerates with cobbles and pebbles of sandstone, granite, quartzite and limestone.

In eastern Nepal, the local surface geology at the town of Charikot (figure 9) consists of the Ulleri Formation (Upper Pre-Cambrian to Late Palaeozoic age) and the Ranimatta and Seti Formations. The Ulleri Formation consists of biotite- and muscovite-containing augengneiss and feldspathic schists. The surface geology at the town of Manthali (figure 9) contains both the Galyan Formation (Upper Pre-Cambrian to Late Palaeozoic age) and the Ranimatta/Seti Formation. The lithology of the Galyan Formation is described as slates, intercalated with thin calcareous slates and carbonates. Locally thick beds of siliceous dolomites are present (HMH, n.d.).

The local surface geology of the Arthunge Municipality (figure 9) contains both the Seti Formation and the Kushma Formation (Upper Pre-Cambrian to Late Palaeozoic age). The Kushma Formation is characterized by massive quartzite, intercalated with phyllites (HMH, n.d.).

Table 8: Surface geology of the studied arsenic affected locations in Nepal.

Location	District	Surface geology	Sediments and rock characteristics
Terai	See table 3	Alluvium, Upper, Middle and Lower Siwalik	Gravels, sands and clays. Conglomerates, sandstones, clays and shales
Kathmandu Valley	Kathmandu, Lalitpur and Bhaktapur	Chandragiri Formation, Tistung Formation	Limestone, quartzite. Phyllite, sandstone and limestone
Pokhara Valley	Kaski	Seti Formation, Ranimatta Formation	Sandstones, gritstones with conglomerate, phyllite and quartzite.
Mustang Valley	Mustang	Higher Himalayan, Thakkhola Formation, Tibetan Sedimentary Zone	Fluvial and fluvio torrential sediments with clay and marlstones. Fluvial and alluvial fan conglomerates with cobbles and pebbles of sandstone, granite, quartzite and limestone. Continental platform sediments with local pro-delta facies, glauconite shales, slates, sandstones and limestones.
Charikot and Manthali	Dolkha and Ramechap	Seti Formation, Ranimatta Formation, Ulleri Formation, Galyan Formation	Sandstones, gritstones with conglomerate and quartzite. Gneiss and schist. Slates, carbonates and dolomites
Arthunge	Myagdi	Seti Formaion, Kushma Formation	Sandstones, gritstones with conglomerate, phyllite and quartzite. Quartzite with phyllite

Northeastern India

As the state of Assam is largely made up of the floodplain of the Lower BRB, most of the identified arsenic groundwater affected districts in Assam are associated with fluvial sediments (table 9). These fluvial sediments have a similar composition as the Newer and Older alluvium of the GRB and consist mostly of sand, silt and clay. A different geological environment to the other indicated districts of Assam, is present in the southern district of Cachar, which mainly has a rock type of siltstone, shale, coal seam and sandstone, belonging to the Barail Group (Oligocene age). Furthermore, a different geological setting is also present in the district of Nagaon, besides the abundant fluvial sediments. A rock type of quartzite, phyllite and schist of the Shillong Group (mid-Cretaceous age) is present here (Verma et al., 2016).

For the district of Thoubal, which was the only arsenic groundwater affected district indicated in the state of Manipur, the geological environment mainly consists of the Disang Group, which has a rock type of ophiolitic rocks (locally known as the Nagaland-Manipur ophiolites having an early Cretaceous age) (Singh et al., 2016; Ovung et al., 2017).

Considering the state of Arunachal Pradesh, the identified arsenic groundwater affected districts of Papum Pare, East Kameng and Lower Subansiri appear to be all associated with the Bomdila Group, regarding their geological settings. The Bomdila Group (Paleoproterozoic age) consists of the rock types schists, phyllite, metavolcanics and quartzite. The district of West Kameng consists of a slightly different rock type of phyllite, quartzite and mica schist belonging to the Dirang Formation (Paleoproterozoic to Mesoproterozoic age) as well as gneiss and high-grade schist (which form the Seta Group). The Dibang Valley area also contains the Bomdila Group, as well as granite, diorite, granodiorite, leucogranite and tonalite of the Lohit Granitoid Complex (Late Cretaceous to Paleocene – Eocene age). In the Tirap district, a rock type of sandstone and shale is present, which are part of the Diasang Group (Eocene age) (Verma et al., 2016).

In the state of Tripura, the geological environment is roughly the same for all the identified arsenic groundwater affected districts. The rock type of the districts consists of sandstone with conglomerate and shale of the Dupi Tila Formation (Plio-Pleistocene age), siltstone, shale, clay and conglomerate belonging to the Tipam Formation (Late Miocene to Early Pliocene age) as well as siltstone, mudstone, sandstones and shales, which make up the Shurma Group (Upper Oligocene to Miocene age) (Verma et al., 2016; Roy et al., 2012; Bharali et al., 2017).

The arsenic groundwater affected districts of Mokok Chong and Mon indicated in the state of Nagaland have a similar geological environment, as both districts are associated with the Barail Group and Disang Group (Verma et al., 2016).

Table 9: Surface geology of the studied arsenic affected states in North-eastern India.

State	Surface geology	Sediments and rock characteristics
Assam	Barail Group, Fluvial sediments, Shillong Group	Siltstone, shale, coal seam and sandstone Sand, silt and clay Quartzite, phyllite and schist
Manipur	Disang Group	Ophiolites
Arunachal Pradesh	Bomdila Group Dirang Formation, Seta Group, Lohit Granitoid Complex, Disang Group	Schist, phyllite, metavolcanics and quartzite Phyllite, quartzite and mica schist. Gneiss and high grade schist Granite, diorite, granodiorite, leucogranite and tonalite Sandstone and shale
Tripura	Dupi Tila Formation, Tipam Formation, Shurma Group	Sandstone with conglomerate and shale. Siltstone, shale, clay and conglomerate. Siltstone, mudstone, sandstones and shales
Nagaland	Barail Group, Disang Group	Siltstone, shale, coal seam and sandstone. Sandstone and shale

Areas having arsenic groundwater concentrations only < 10 µg/L

Uttarakhand

In the state of Uttarakhand (table 10), the surface geology solely consists of the Jaunsar Group (Neoproterozoic age) in the districts of Dehradun, Pauri and Uttarkashi (Das & Modak, 2019). This formation is also present in the districts of Tehri, Chamoli, Nainital and Champawat and mainly consists of quartzites, limestones, slates and phyllite. The upper part of the Jaunsar Group (known as the Nagthat Formation) also contains conglomerates, arkoses, grits, quartzites and sandstones (Dhital, 2015).

As the districts of Haridwar and Udham Sing Nagar are part of the Gangetic floodplain, the local surface geology of these districts consists of the Newer and Older alluvium.

The local surface geology of the Tehri district is characterised by the Rautgara Formation (Mesoproterozoic age) in addition to the Jaunsar Group. The Rautgara Formation is composed of calcareous slates, mylonites and coarse-grained quartz arenites (Joshi, 2013). This formation also occurs in the district of Rudraprayag.

The surface geology of the Chamoli district also includes the Berinag and Bajinath Formations (Proterozoic to Palaeozoic age). The Berinag Formation mostly consists of schistose quartzite (Bose & Mukherjee, 2019), whereas the Baijnath Crystalline is characterized by quartzites and chlorite schists (Chamyal, 1991).

Another distinctive geological feature present in Uttarakhand is the Almora-Ramgarh Group (Paleoproterozoic age), which is found in the southeastern part of the state (Nainital, Champawat and Almora districts). The Almora Group is composed of coarse-grained mica-schist and micaceous quartzite and granite (Rawat, 2011; Joshi et al., 2016).

Regarding their surface geology, the districts of Pithoragarh and Bageshwar contain the Berinag Formation, as well as the Pithoragarh Formation (Mesoproterozoic age). The Pithoragarh Formation consists of phyllite, cherty quartzite, shale, dolomite and limestone (GSI, 2012).

Table 10: Surface geology of districts in Uttarakhand having arsenic groundwater concentrations only < 10 µg/L.

District	Surface geology	Sediments and rock characteristics
Dehradun, Pauri, Uttarkashi	Jaunsar Group	Quartzites, limestones, slates and phyllite
Haridwar and Udham Sing Nagar	Newer alluvium + Older alluvium	Sand (locally micaceous), silt, clay and gravel
Tehri	Jaunsar Group, Rautgara Formation	Quartzites, limestones, slates and phyllite.
Chamoli	Jaunsar Group, Berinag Formation, Baijnath Crystalline	Quartzites, limestones, slates and phyllite. Quartzites Quartzites and schists
Rudraprayag	Rautgara Formation	Quartz arenites, slates and mylonites
Nainital and Champawat	Jaunsar Group, Almora-Ramgarh Group	Quartzites, limestones, slates and phyllite. Schist, quartzite and granite
Almora	Almora-Ramgarh Group	Schist, quartzite and granite
Pithoragarh and Bageshwar	Berinag Formation, Pithoragarh Formation	Quartzites phyllite, quartzite, shale, dolomite and limestone

Sikkim
As the literature unfortunately did not mention the specific monitoring locations in the state of Sikkim, the local surface geology that it is associated with these monitoring locations is difficult to point out. Hence, a broad overview of the surface geology is given.

The southern part of Sikkim is mainly composed of chlorite-sericite schist, mica schist, biotite phyllite, quartzite and slates, with locally mylonitic granite gneiss. Sandstone and carbonaceous shale with coal also occur locally. The remaining surface geology mostly consists of migmatite, augen gneiss with kyanite, leucogranite, sillimanite with kyanite, biotite gneiss and sillimanite granite gneiss (Baruah et al., 2019).

Mizoram
Similar to the situation in Sikkim, the local surface geology of Mizoram is also difficult to point out, as the literature did not give notice of specific monitoring locations. Therefore, the surface geology of Mizoram is also given as a broad overview. The surface geology of Mizoram is composed of the Shurma Group and Tipam Formation (Bharali et al., 2017), as mentioned in table 9.

Nepal
The Badigad catchment area has a surface geology composed of the Siuri (Pre-Cambrian age), Lakharpata and Galyang (both Upper Pre-Cambrian to Late Palaeozoic age) Formations (table 11). The Siuri Formation consists of muscovite, biotite and garnetiferous schists, quartzites and mylonitic augen gneiss. The Lakharpata Formation is characterized by fine grained limestones and dolomites with shales and quartzites (HMH, n.d.).

The local surface geology at Dhankuta Municipality is characterized by the Himal Group (Pre-Cambrian age), which is mainly composed of garnet gneisses (containing kyanite and biotite), garnetiferous mica schists, quartzites and marbles (HMH, n.d.).

The Marsyangdi catchment area contains the Sombre Formation (Silurian – Devonian age), Annapurna Formation (Cambrian – Ordovician age) and units 1, 2 and 3 of the Greater Himalayan Sequence (locally of Cambrian – Ordovician age) (Ghezzi et al., 2019). The Sombre Formation mostly contains gritty dolomites, shales and sandstones, while the Annapurna Formation is composed of sandstone, siltstone, and limestone (Neupane et al., 2018). The geology of the Greater Himalayan Sequence is categorized into 3 different units for the area of the Marsyangdi catchment: unit 1 is mostly made up of calc-silicate, quartzite, mica schist, paragneiss and aluminosilicate bearing migmatite; unit 2 consists of quartzite and calc-silicate and marble-bearing rocks; and unit 3 is mainly composed of orthogneiss and metapelite bearing mica, sillimanite and garnet (Walters & Kohn, 2017; Ghezzi et al., 2019).

The local surface geology in the Jhikhu Khola catchment area is characterized by the Upper Nawakot Complex (Pre-Cambrian to Lower Palaeozoic age), with mostly local alluvial deposits, phyllite, mica schist and quartzite (Dongol et al., 2005; Nakarmi, 2000).

Table 11: Surface geology of locations in Nepal having arsenic groundwater concentrations only < 10 µg/L.

Location	**District**	**Surface geology**	**Sediments and rock characteristics**
Badigad catchment	Gulmi and Baglung	Siuri Formation, Lakharpata Formation Galyang Formation	Schists, quartzites and mylonitic gneiss. Limestones, dolomites, shales and quartzites. Slates, carbonates and dolomites.
Dhankuta Municipality (Koshi catchment)	Dhankuta	Himal Group	Gneisses, schists, quartzites and marbles
Marsyangdi catchment	Manang and Lamjung	Sombre Formation, Annapurna Formation, Greater Himalayan Sequence	Dolomites, shales and sandstones. Sandstones, siltstones and limestones. Calc- silicate, quartzite, mica schist, paragneiss, migmatite, marble, orthogneiss and metapelite
Jhikhu Khola catchment	Kavrepalanchok	Upper Nawakot Complex	Alluvial deposits, phyllite, mica schist, quartzite

Differences in surface geology

In general, there does not seem to be a clear difference in terms of surface geology (and associated sediments and rock characteristics) between the areas containing arsenic anomalies and areas having low arsenic groundwater concentrations. It appears that there is not simply a single formation or rock type which acts as a main primary source of arsenic. This depicts a highly nuanced and complex situation in the upstream areas (Himalaya) of the GRB and Indian BRB, where multiple possible sources contribute to arsenic groundwater enrichment.

Indicating possible arsenic sources

This section aims to correlate the arsenic groundwater anomalies with the associated surface geology and to discuss possible (primary) sources of arsenic as found as natural pollution in the GRB and Indian BRB.

The arsenic groundwater anomalies in the Indian states of West Bengal, Uttar Pradesh, southern Uttarakhand (Haridwar district) and the Terai region in Nepal are mainly or partly located in the alluvial plain of the Ganges River. Although some of the arsenic groundwater anomalies occur at locations with a different surface geology (porphyritic granite and leucogranite of the Bundelkhand Granitoid Complex in Uttar Pradesh and sandstone, limestone and dolomite of the Vindhyan Supergroup in Bihar), these are only very localized and the vast majority of the anomalies is associated with the Older and Newer alluvia of the Ganges River. Since this study has indicated several locations of arsenic groundwater anomalies in the upper reaches of the Ganges, namely the various catchment areas in the Nepalese Himalayas, it is highly unlikely that the primary source which causes the natural arsenic groundwater pollution in the GRB can be found in the alluvial plains of West Bengal, Bihar, Uttar Pradesh, Uttarakhand and the Nepali Terai.

The indication of arsenic groundwater anomalies in different catchment areas in the Nepalese Himalayas also advocates against the Siwaliks of being the primary source of natural arsenic contamination in the GRB, which is in line with the ideas of Mukherjee er al. (2014). This, however, does not disregard the Siwaliks of being a significant (secondary) source of arsenic and should not be overlooked as a threat for local enriched arsenic groundwater. As many studies have pointed out: the Siwaliks supply a significant amount of arsenic-rich sediments to the Terai, which has led to elevated arsenic groundwater concentrations in most of the Nepali districts of the Terai (e.g. Chakraborty et al., 2015; Guillot et al., 2015; Jain et al., 2022).

When looking at the arsenic anomalies in Pokhara Valley, Arthunge Municipality and the area near the towns of Charikot and Manthali, the Seti, Ranimatta and Ulleri Formations appear to be abundant at these locations. These formations are not present in Badigad catchment, where arsenic groundwater concentrations were found to be low. Since the Seti Formation and the Ranimatta Formation show a similarity to each other in terms of their rock characteristics and seem to be intertwined as presented in (HMH, n.d.), these formations will be further interpreted as having the same lithology. In terms of mineralogy, the Seti/Ranimatta and Ulleri Formations are characterized by the presence of biotite and muscovite. As pointed out by Chakraborty et al. (2007) and Seddique et al. (2008), silt-sized micas, especially biotite and muscovite, can act as an effective adsorption site for arsenic and hence weathering of such minerals is a mechanism that significantly contributes to local arsenic-enriched groundwater concentrations. However, the high sorption capacity of arsenic to biotite and muscovite does not indicate that these minerals (and therefore the Seti/Ranimatta and Ulleri Formations) are the primary sources responsible for the arsenic groundwater contamination. Although these minerals and associated formations can and should be regarded as a significant (secondary or intermediary) source of arsenic, it does not rule out that the primary source of arsenic can be found at an higher altitude. Therefore, it is important to focus on arsenic groundwater anomalies found in areas located higher up in the Nepalese Himalayas.

The Mustang Valley in the Mustang District is regarded as one of the most high-altitude (roughly 3500 m.a.s.l) and remote districts of the Nepalese Himalayas and is located in the Western Development Region (Fort, 2015). Since arsenic groundwater concentrations of well above 50 µg/L (to a maximum of 436 µg/L) have been found here, this also indicates that a primary source of arsenic may be found in the vicinity of the Mustang Valley. When focussing on the mineralogy of the surface rocks, it can be pointed out that the mineral glauconite is present in the Himal Group, located in the Mustang Valley. This mineral was previously not observed in any other lithologies of surface rocks, associated with arsenic groundwater anomalies in the GRB. Glauconite-bearing sediment is known to be able to release arsenic into local groundwater, as indicated by studies in Belgium (Cappuyns et al., 2002) and New Jersey (Mumford et al., 2012). However, although glauconite-bearing deposits have a widespread occurrence, relatively few studies have been focussed on the arsenic content of glauconitic sediments (Barringer et al., 2011). Therefore, it cannot be concluded that this glauconite-bearing deposit of the Himal Group is the (only) primary source of arsenic in the Mustang Valley and it is necessary to look at other possible additional primary arsenic sources in the area.

In the research carried out by Emerman et al. (2014), the authors categorized the arsenic groundwater concentrations in the Mustang Valley into two regions, whereby region 1 was indicated as having high arsenic groundwater concentrations and region 2 as having low arsenic groundwater concentrations anywhere. It was found that region 1 received sediment input from the local Mustang and Mugu Granites, whereas region 2 did not. This led to the believe that weathering of these local granites was likely the origin of arsenic in the groundwater. This was later confirmed by Ghezzi et al. (2017), who also found that weathering of the outcropping granitic rocks in the Mustang Valley is likely to be the primary source of arsenic in the region.

Leucogranites as possible primary arsenic source in the GRB

The aforementioned Mugu and Mustang granites belong to a series of leucogranites (Tethyan Himalayan leucogranites) in Nepal and southern Tibet (figure 13) which are part of an extensive igneous province in the Himalayan orogen and have an age of Late Oligocene-Miocene (Guo & Wilson, 2012). Since these leucogranites are characterized as being rich in tourmaline (Dhital, 2015; HMH, n.d.), there might be a connection between the weathering of this mineral and elevated arsenic concentrations in groundwater. However, contradicting this idea is the absence of high arsenic

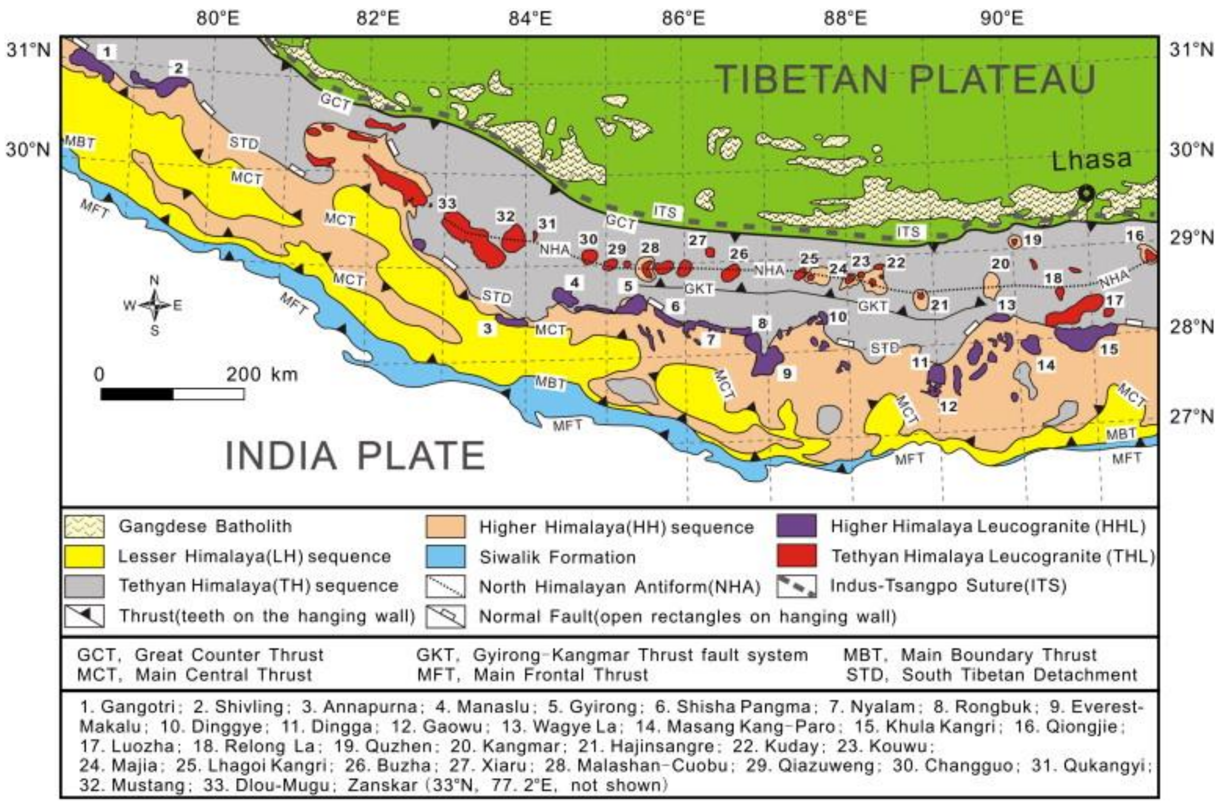

Figure 13: Location of the Higher Himalayan- and Tethyan Himalayan leucogranites (Guo & Wilson, 2012).

groundwater concentrations in the state of Uttarakhand as found in this study. Leucogranites, particularly those rich in tourmaline, are also present in this state (Searle et al., 1993). Hence, if (weathering of) tourmaline would be the key mineral responsible for arsenic release, elevated arsenic groundwater concentrations would also be expected in Uttarakhand. Moreover, the state of Sikkim also contains tourmaline leucogranites in its Himalayan regions (Guo & Wilson, 2012). However, this state (and neighbouring eastern Nepal as indicated by the Dhankuta Municipality) also appears to not have any problems with elevated arsenic groundwater concentrations. Nevertheless, this does not disregard the leucogranites as being a possible primary source of arsenic. In fact, the Manaslu leucogranite, which has a high similarity with the Mugu and Mustang leucogranites (Le Fort & France-Lanord, 1995), was also assumed by Mueller (2018) of being the original source of natural arsenic pollution in Nawalparasi. Unfortunately, the composition of the leucogranites in terms of arsenic content is unknown to the best of my knowledge.

Leucogranites as indicated by trace elements

According to Mueller (2018), the trace elements Li, B, P, Mn, Zn, As, Sr, Pb and U can be used as indicators of a leucogranite source rock. Therefore, elevated concentrations of these trace elements in rivers flowing downstream from the Nepalese Himalayas could hint towards leucogranites as source. The data of three geochemical studies (Pant et al., 2020; Paudyal et al., 2016; Ghezzi et al., 2019) are presented in table 12, with focus on the trace elements associated with leucogranites and the global mean values (Gaillardet et al., 2013) for comparison.

The Gandaki River generally appears to have elevated trace element concentrations, in comparison to the global mean values. As this river originates in the Mustang Valley, these elevated trace elements concentrations would support the theory of the leucogranites in this region being the primary source of arsenic.

The Indrawati and Dudh Koshi Rivers are tributaries of the Koshi River, located in eastern Nepal. When directly comparing these two rivers to each other, it is observed that the trace elements concentrations for the Dudh Koshi river are generally higher than for the Indrawati river. Hence, this would indicate that the Dudh Koshi has a leucogranitic source, whereas the Indrawati does not. The observation that the Indrawati river does not contain any arsenic, while the Dudh Koshi does have elevated dissolved arsenic, further implies that leucogranites could possibly be a (primary) arsenic source. Focussing on the course of the Indrawati river (HMH, n.d.), this river possibly has (had) a significant contribution to the (alluvial) sediments deposited in the Jhikhu Khola catchment. The absence of a leucogranitic source of arsenic for the origin of this river and its sediment, may explain that no elevated arsenic groundwater concentrations occur in this catchment.

The Marsyangdi River does not clearly show elevated trace element concentrations, hence not indicating a leucogranitic source. This would be expected as the current study indicated that the Marsyangdi River basin was found to be clear of elevated arsenic groundwater concentrations.

Table 12: Concentrations (in µg/L) of dissolved trace elements in rivers in Nepal, associated with leucogranites, in comparison with the global mean values of these trace elements in river waters (Pant et al., 2020; Paudyal et al., 2016; Ghezzi et al., 2019; Gaillardet et al., 2013).

River	As	Li	B	P	Mn	Zn	Sr	Pb	U
Gandaki (mean)	2.02	60.64	-	-	6.63	127.70	322.48	0.61	4.57
Indrawati (average)	0	1.53	-	-	14.26	7.12	42.29	0.69	0.75
Dudh Koshi (average)	3.68	14.26	-	-	28.24	16.74	12.38	1.82	4.42
Marsyangdi (average)	0.54	12.1	-	-	<0.2	-	291	<0.2	3.9
Global values (mean)	0.62	1.84	-	-	34.0	0.60	60.0	0.08	0.37

Beyond the GRB: leucogranitic arsenic sources in the Indian BRB

The majority of the arsenic groundwater anomalies in the Indian BRB can be linked to the fluvial sediments that are deposited in the floodplain of the Brahmaputra River, which cover most of Assam (and parts of Bangladesh). Similar to the situation in the GRB, the primary source of the natural arsenic groundwater pollution in the Indian BRB is not likely to be located in its floodplain/alluvial plain, since arsenic anomalies have also been indicated at various locations outside these plains in a more mountainous environment.

The theory of leucogranites being the primary source of arsenic could also be linked to some of the arsenic anomalies in the Indian BRB, which appear not to be associated with the sediments of the Brahmaputra River and, hence, have local arsenic sources. The occurrence of (Higher Himalayan) leucogranites in western Arunachal Pradesh (Bhattacharjee & Nandy, 2007) could be linked to the arsenic anomalies in West and East Kameng and Lower Subansiri, whereas the presence of leucogranites in eastern Arunachal Pradesh (Goswami, 2013; Vermi et al., 2016) could be associated with the elevated arsenic groundwater concentrations in the Dibang Valley. However, this study also indicated arsenic anomalies further south in the Indian BRB, in the states of Nagaland and Manipur where no leucogranites seem to exist. Thus, other rocks than leucogranites should be identified as possible sources for these anomalies. Vermi et al. (2016) mentioned the ophiolite and arc-related volcanic rocks of the Indo-Burmese suture zone located in the Naga Hills as being the probable arsenic provenance of the southern part of the Foreland Basin of the Brahmaputra River. This provenance area may also be linked to the arsenic anomalies in Nagaland and Manipur.

Linking to hypotheses

This section targets to test the three main hypotheses of Stanger (2005), Guilliot & Charlet (2007) and Mueller (2018).

The results of this study clearly point towards the hypothesis of Mueller (2018), as arsenic anomalies in the GRB appear to be closely linked to leucogranites. However, leucogranites also occur in areas where only low arsenic groundwater concentrations were found. Moreover, results of this study indicate that regions with an absence of leucogranites also appear to be able to have high arsenic groundwater concentrations. Thus, the leucogranites do not always act as a source for elevated arsenic groundwater concentrations and other (local) arsenic sources need to be considered as well.

Although the hypotheses of Stanger (2005) and Guilliot & Charlet (2007) appear to be not valid for the GRB, in essence, they are not completely rejected by this study, as they could still play a role for the (Indian) BRB. In fact, Verma et al. (2016) indicated that the volcanics and ophiolites of the Indus-Tsangpo suture zone and the ophiolitic and volcanic rocks in the Namcha Barwa syntax, could act as probable arsenic provenances for the northern region of the Foreland Basin, belonging to the Brahmaputra River. Furthermore, studies indicated that elevated arsenic concentrations occur in the soil of the southern and eastern Lhasa Terrane in Tibet (Sheng et al., 2012) and that the Yarlung Tsangpo (local name for the Brahmaputra River in Tibet), which cuts through the Lhasa Terrane, might be vital in contributing to the increased arsenic groundwater concentrations in the Indian BRB (Li et al., 2011). Therefore, the arsenic sources as mentioned by Stanger (2005) and Guillot & Charlet (2007) may not act as the primary source of arsenic in the GRB, yet still contribute (to a certain extent) to the natural arsenic groundwater contamination in the Indian BRB.

Suggestions for further research

Since this study was limited by the means of time and resources, various further research is recommended:

- So far, no studies concerning arsenic groundwater concentrations in western Nepal (except the western Terai region), Bhutan and southern Tibet have been conducted. Thus, arsenic groundwater testing is asked for these regions.
- Since weathering of the leucogranites in Uttarakhand and Sikkim appears to not cause local elevated arsenic groundwater concentrations, a study focusing on the comparison (in terms of mineral composition) between these leucogranites and the leucogranites in Central Nepal is needed.
- The results of this study (arsenic anomalies distribution and concentrations) may be used for drinking water contamination risk analysis.
- To confirm whether the arsenic anomalies in Arunachal Pradesh may actually be associated with the leucogranites in that state, a geochemical study focusing on the leucogranitic trace elements is needed.
- An investigation about the sediment budgets and fluxes at annual and geological time scale between the Himalayan leucogranites and the Indo-Gangetic Plain, may imply if this source is realistic in terms of volume and sediment fluxes.

Limitations

In general, the present study was heavily depending on and limited by the quality and quantity of data that was able to be accessed. As table 1 shows, some reports contained significantly less samples compared to others. Also some reports did not clarify the amount of samples taken or failed to show the exact sample locations. Furthermore, there were contradicting data at times, where studies conducted in the same area, locally had significantly different arsenic groundwater concentrations (this occurred in the Haridwar district and the Mustang Valley). This also applied to geological maps from different studies which showed some variations in the surface geology for the same area. Moreover, the data of the arsenic groundwater concentrations also differed in terms of showing maxima, averages, mean values or actual single values.

Collecting data of higher elevated and remote regions in Nepal faced difficulties, as the vast majority of arsenic groundwater studies in Nepal were focused on the Terai. As a result, only few arsenic groundwater studies were found in elevated and remote regions of Nepal. None of the approached institutes in Nepal gave access to the raw data of arsenic groundwater concentrations, thus for the Terai, only general distribution maps showing arsenic groundwater concentrations were available.

6. Conclusions

Arsenic groundwater contamination poses serious health issues in the Ganges and Indian Brahmaputra River Basins and other areas worldwide, whereby the current occurring arsenic contamination in the Indian subcontinent is regarded as "the largest mass poisoning of a population in history". Hence, it is crucial to find the primary arsenic sources which cause the natural groundwater pollution in the downstream deltaic areas of the Ganges River and Indian Brahmaputra River Basins.

The results showed that both the Ganges and the Indian Brahmaputra River Basins are affected by high arsenic groundwater concentrations. In the GRB, arsenic groundwater anomalies have been found in the Indian states of West Bengal, Bihar, Uttar Pradesh and southern Uttarakhand, as well as the Terai region and various catchment areas in Nepal, whereas arsenic groundwater anomalies in the Indian BRB have been found in the states of Assam, Manipur, Arunachal Pradesh, Tripura and Nagaland. Areas having only low arsenic groundwater concentrations were found in the Indian states of Uttarakhand, Sikkim and Mizoram as well as some catchment areas in the Nepalese Himalayas.

The current study also found that there were no clear correlations between the occurrence of high concentrations of heavy metals associated with sulphides (Fe, Cu, Pb, Co, Zn and Ag) and the distribution of arsenic groundwater anomalies. Hence, the occurrence of heavy metal anomalies cannot be used as proxy for the source of arsenic. When focussing on the surface geology and associated sediments and rock characteristics, it appeared that there is not simply a single formation or rock type which acts as a main primary source of arsenic. This depicts a highly nuanced and complex situation in the upstream areas (Himalayas) of the GRB and Indian BRB, where multiple possible sources contribute to arsenic groundwater enrichment.

In the Nepalese Himalayas, the Seti, Ranimatta and Ulleri Formations seem to be acting as a source of arsenic release, whereby the weathering of surface rocks containing biotite and muscovite is a mechanism that possibly contributes significantly to local arsenic-enriched groundwater. The leucogranites of the Tethyan Himalayas probably act as a primary source of arsenic, as weathering of these rocks appear to be causing arsenic groundwater enrichment in some high-altitude and remote districts (e.g. Mustang Valley) within the Nepalese Himalayas. Therefore, the findings of this study are in line with the hypothesis of Mueller, (2018), who stated that leucogranites of the Nepalese Himalayas could be the primary source of arsenic in the GRB. However, leucogranites also occur in areas where only low arsenic groundwater concentrations have been found (Sikkim and Uttarakhand). Moreover, results of this study indicated that regions with an absence of leucogranites, such as in the Indian states of Nagaland and Manipur, also appear to be able to have high arsenic groundwater concentrations. Thus, the leucogranites are not the single source for elevated arsenic groundwater concentrations and other (local) yet-unknown arsenic sources need to be considered as well.

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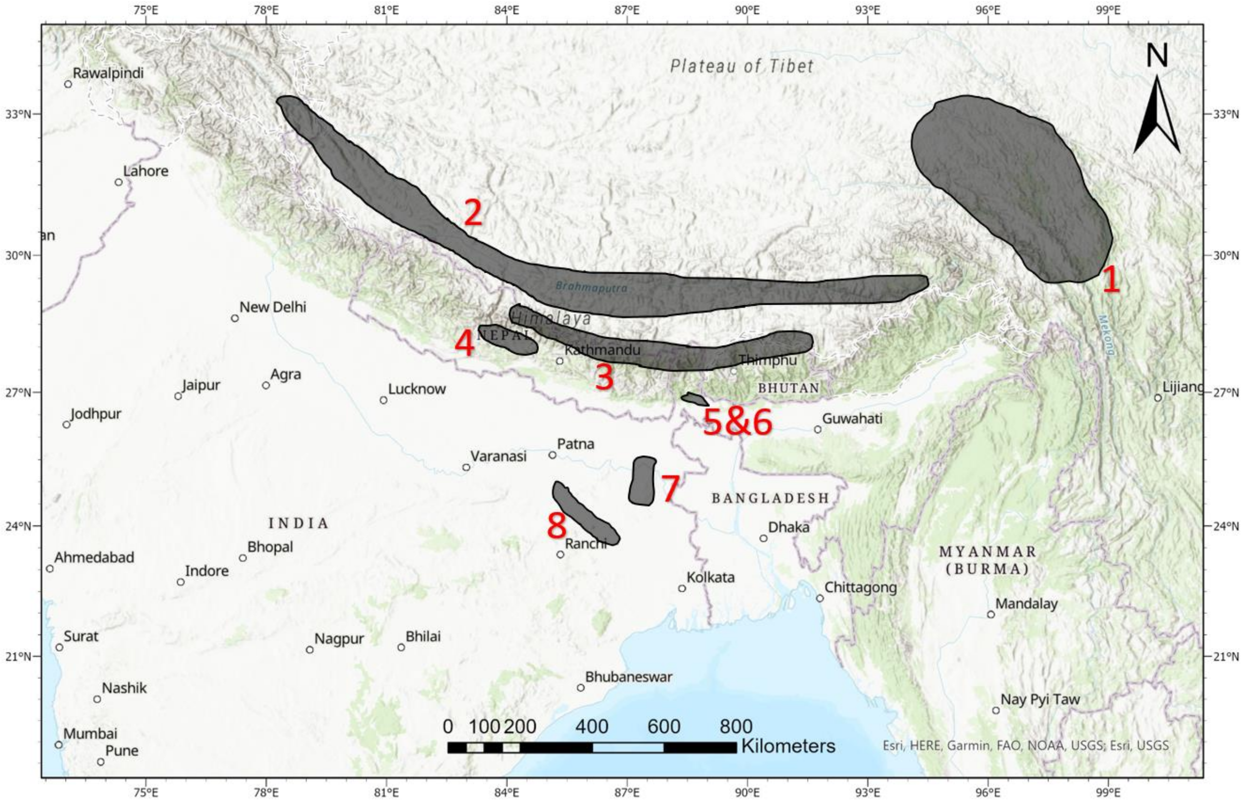
Appendices

Appendix A: Potential arsenic sources

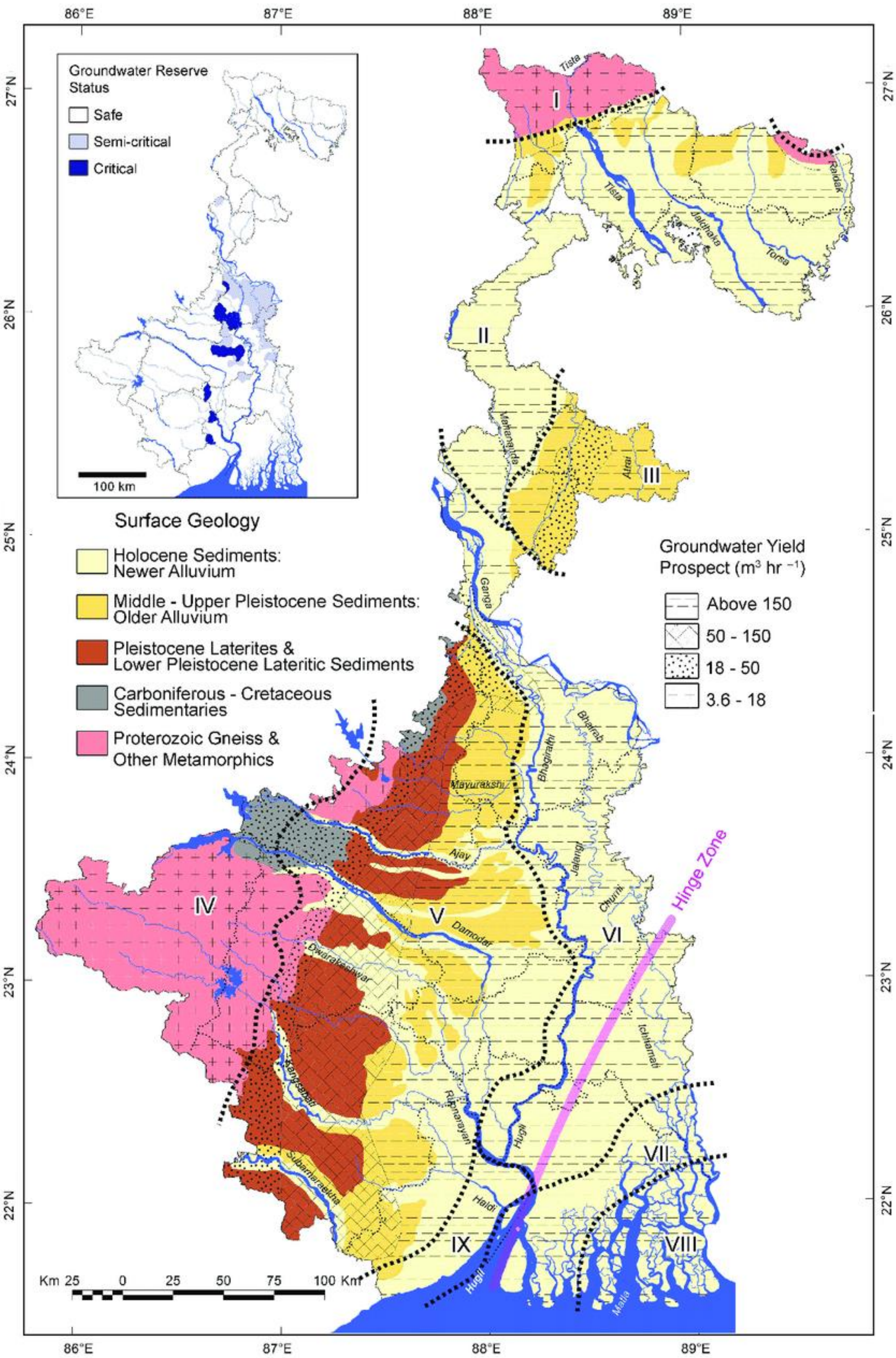
A1: Geospatial map showing the potential arsenic sources found in the literature



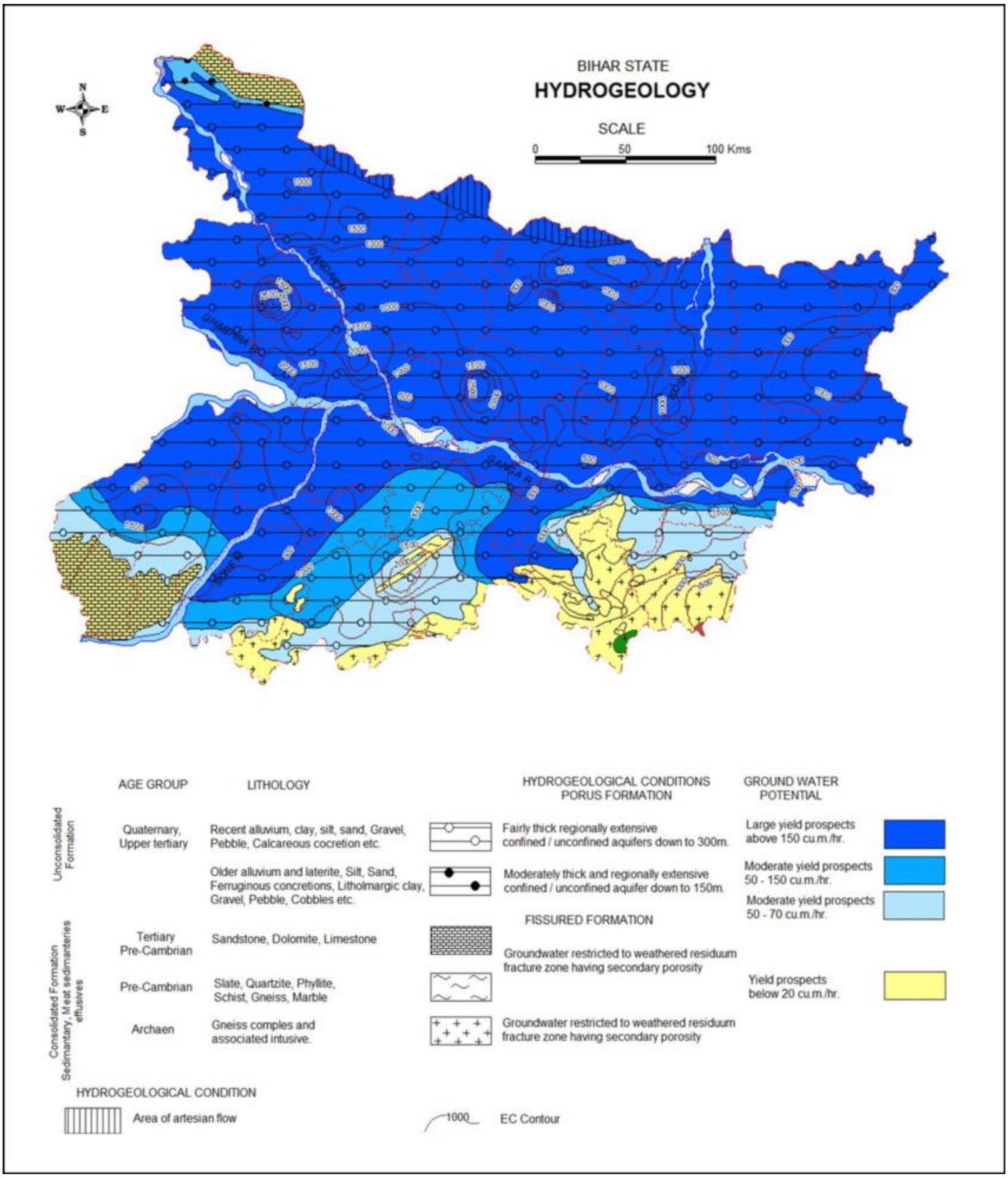
1: Quamdo-Simao volcanic and ophiolite province, 2: Indus-Tsangpo suture zone, 3: Higher Himalayan leucogranites, 4: Black schists of the lesser Himalayas, 5: Isolated sulphide outcrops in the Darjeeling Himalayas, 6: Gorubathan base-metal deposits, 7: Gondwana coal seam in the Rajmahal Traps, 8: Bihar mica-belt.

Appendix B: Geological maps

B1: Geological map of West Bengal

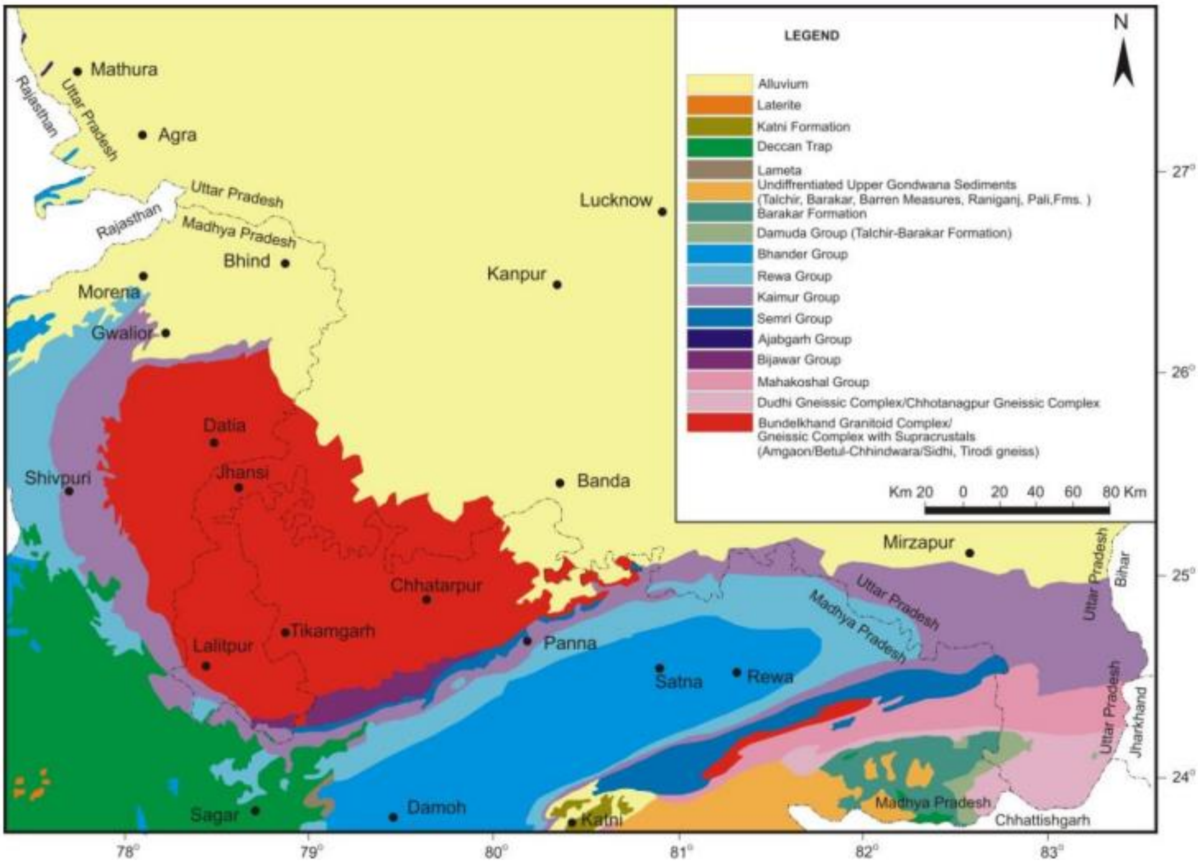


B2: hydrogeological map of Bihar

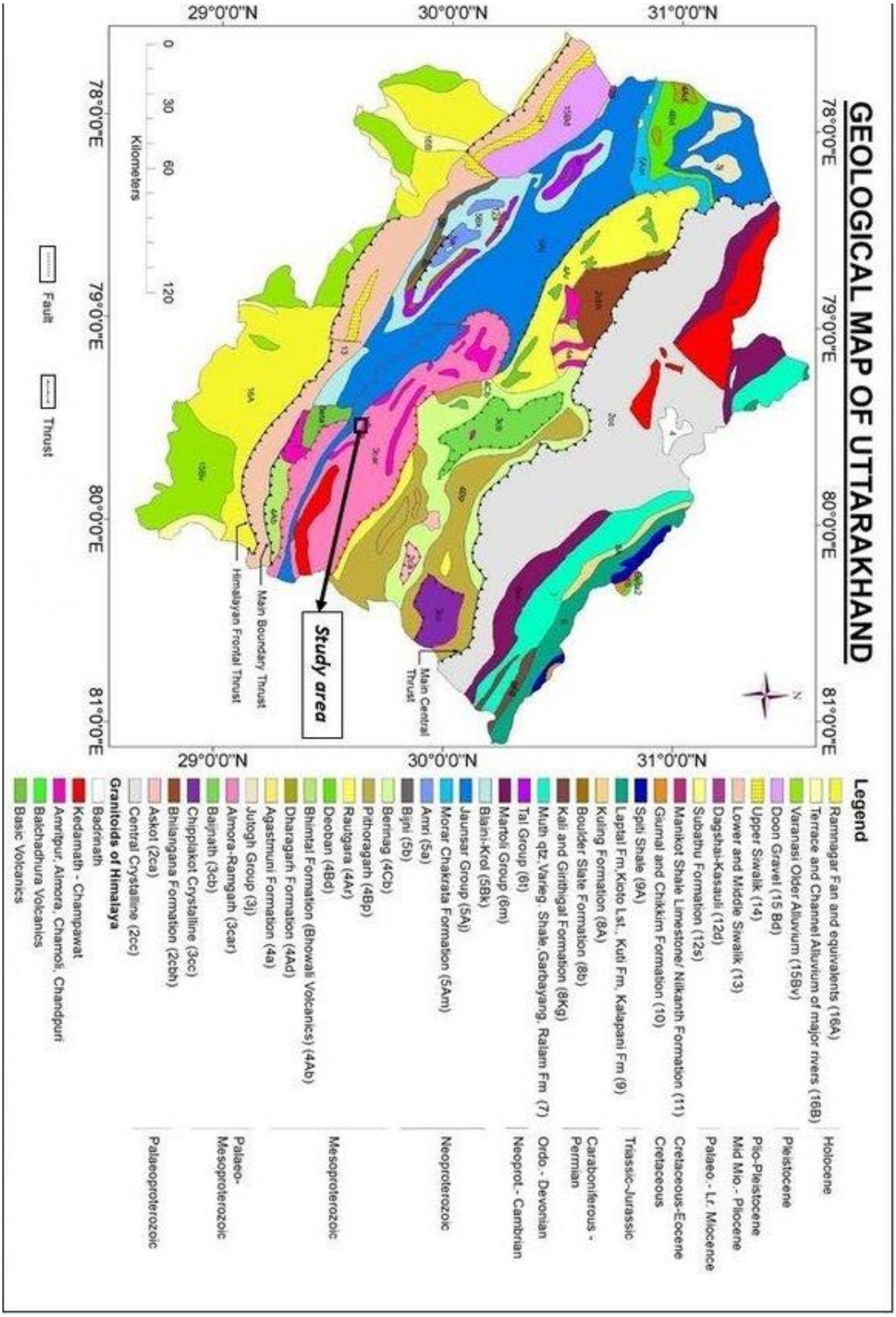


B3-1: Geological map of Uttar Pradesh

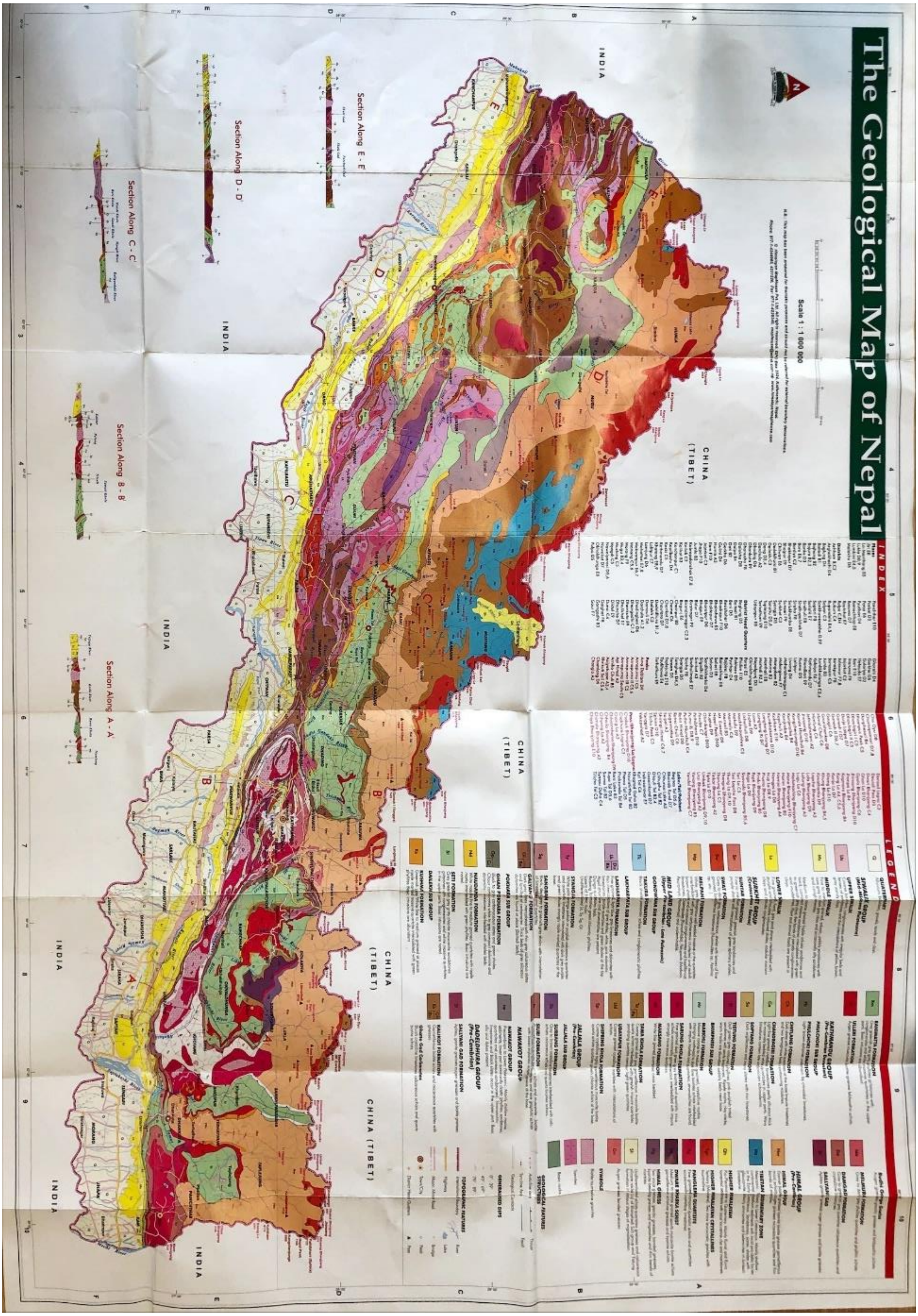
B3-2: Geological map of southern Uttar Pradesh



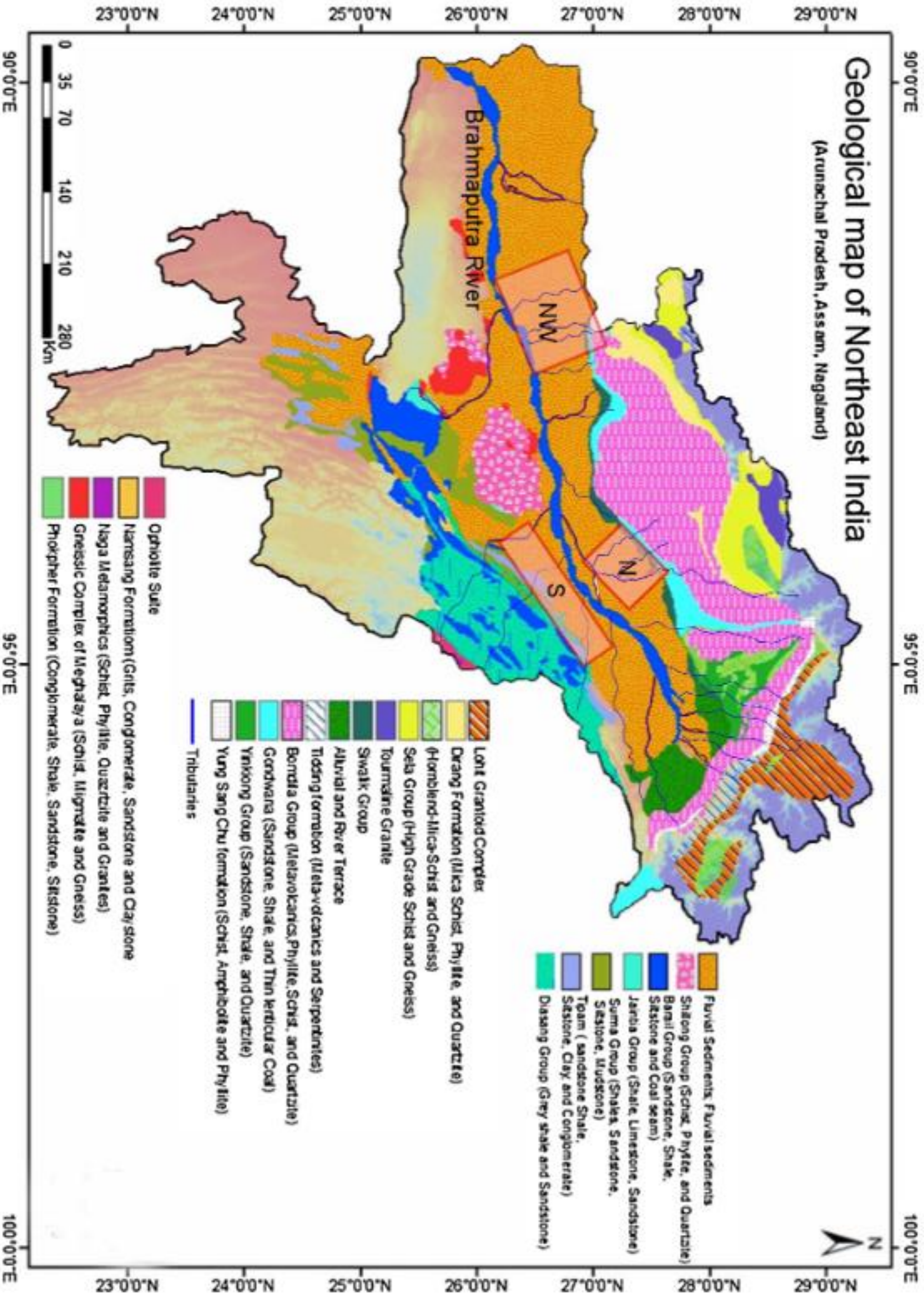
B4: Geological map of Uttarakhand



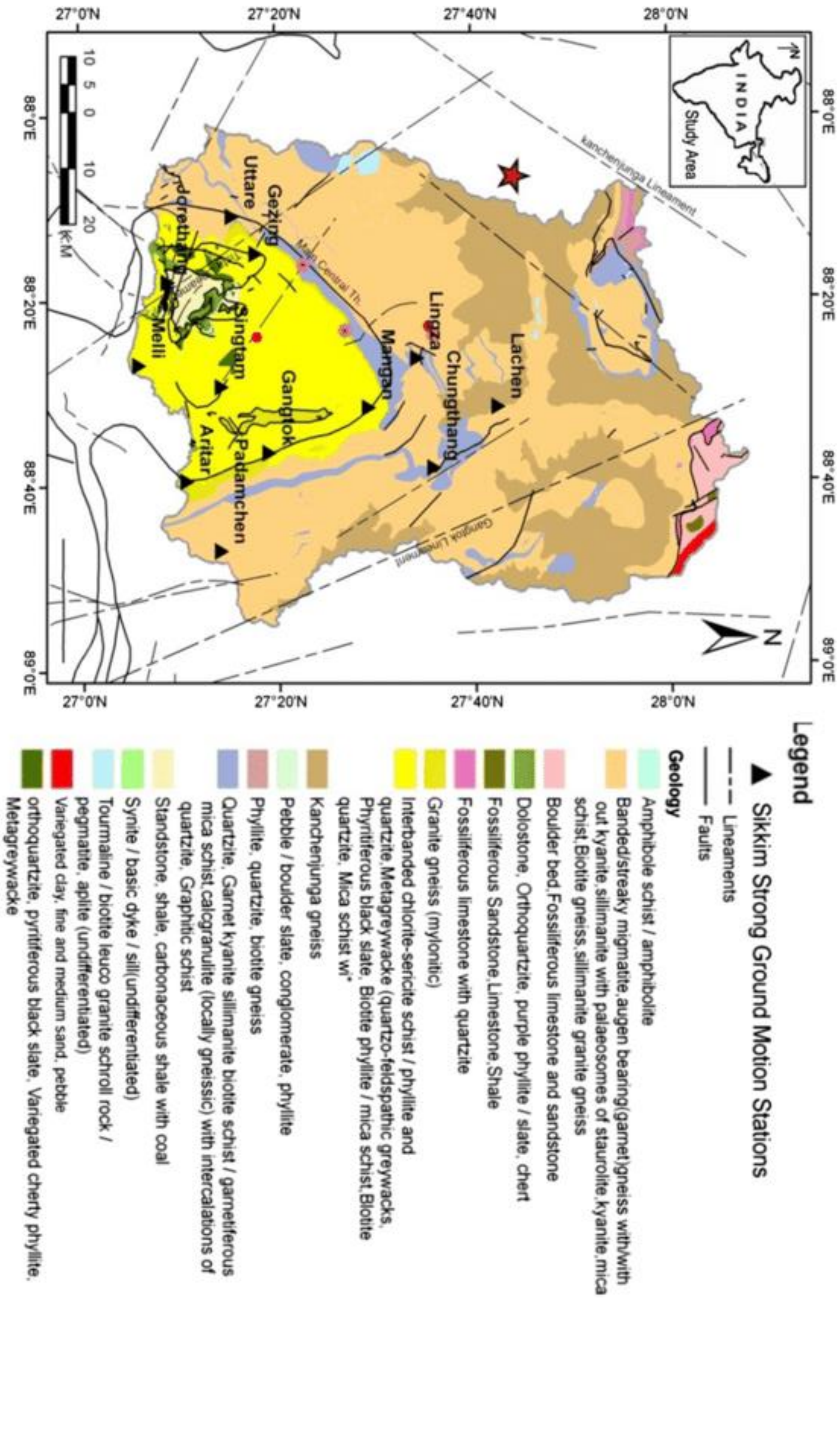
B5: Geological map of Nepal



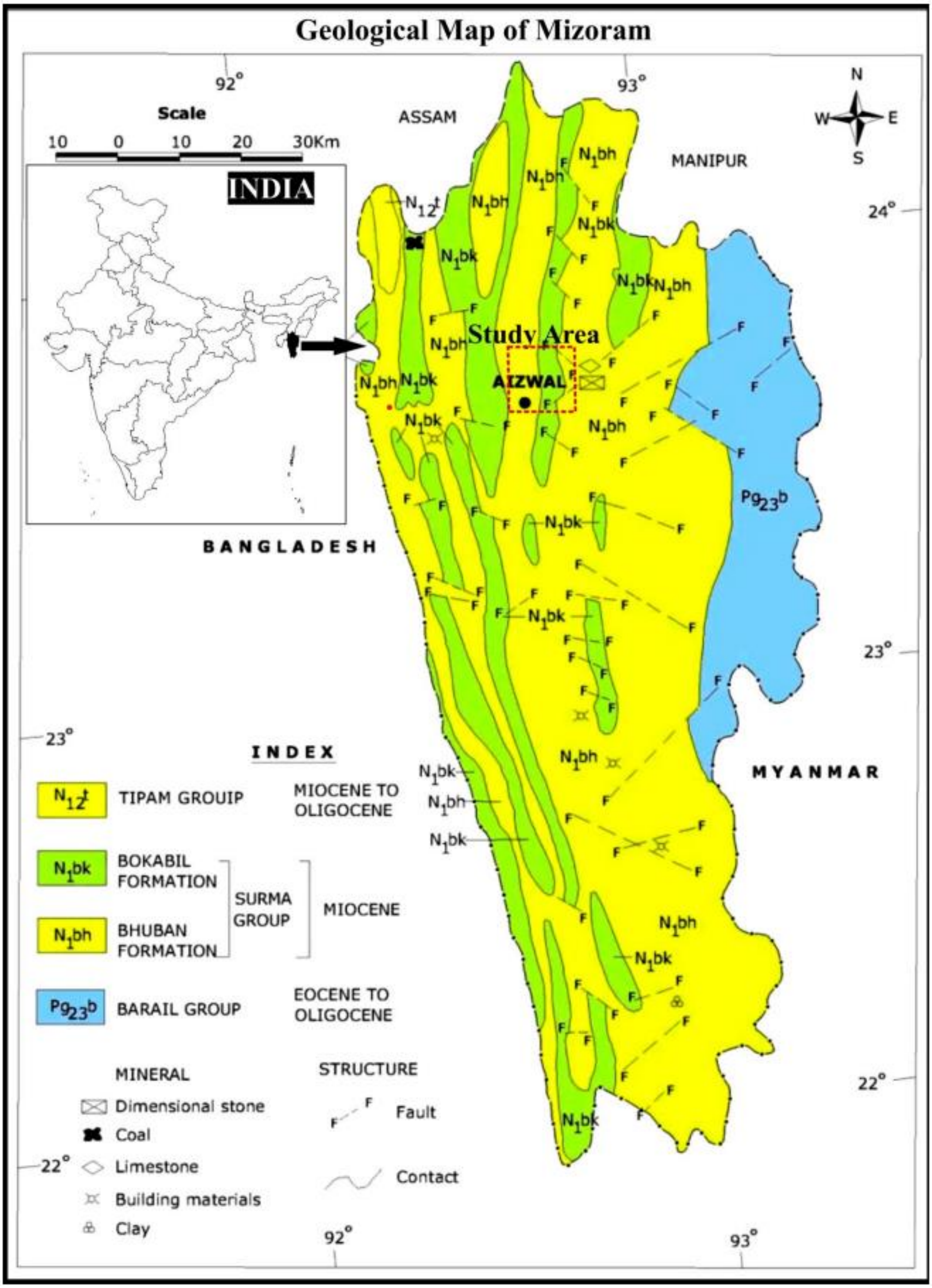
B6: Geological map of Northeast India



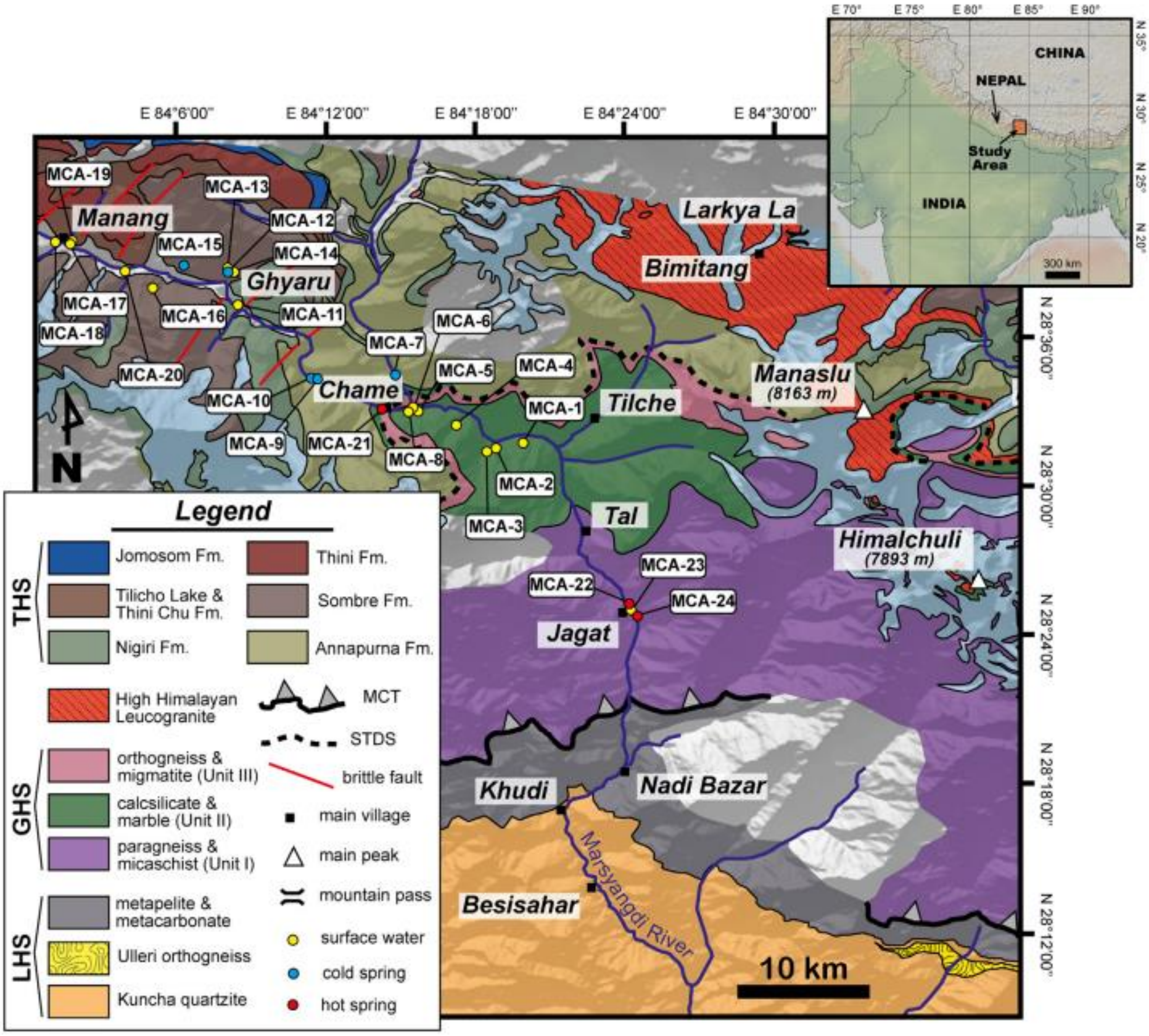
B7: Geological map of Sikkim



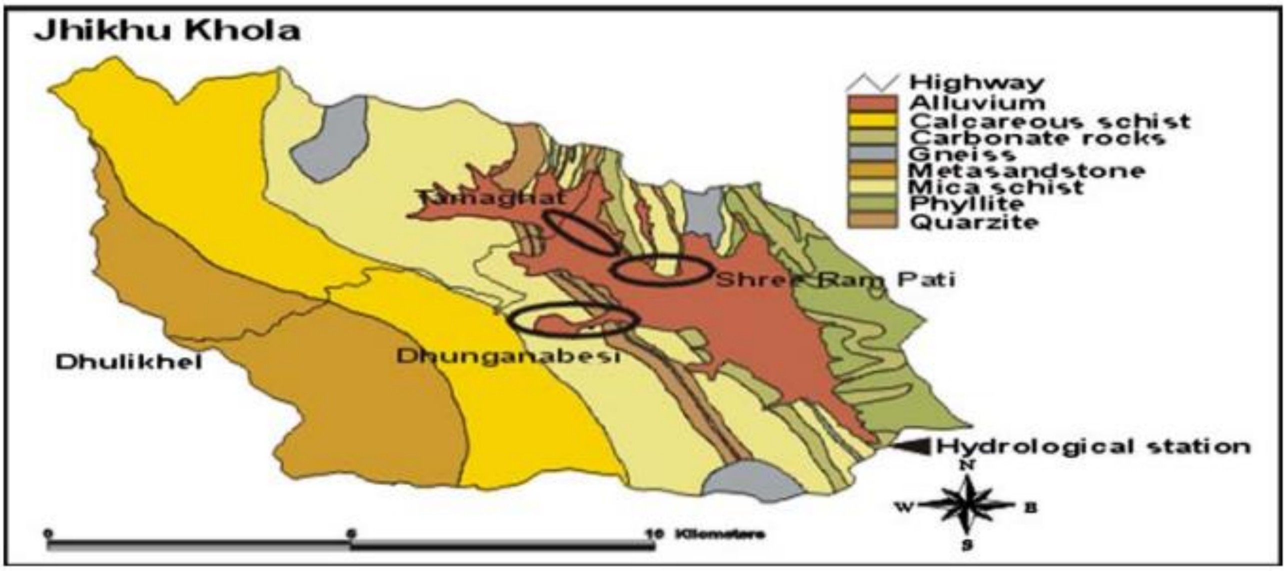
B8: Geological map of Mizoram



B9: Geological map of the Marsyangdi basin



B10: Geological map of the Jhikhu Khola basin



Appendix C: Extended data on arsenic groundwater concentrations

C1: Ranges in elevated (> 10 µg/L) maxima of arsenic groundwater concentrations for districts in West Bengal

District	**As (µg/L)**
Burdwan	10.0 – 50.0
Hooghly	20.0 – 52.0
Howrah	10.0
Kochbihar	40.0
Malda	10.0 - 402
Murshidabad	10.0 - 405
Nadia	10.0 – 332
North 24 Parganas	10.0 – 282
South 24 Parganas	10.0 – 83.0

C2: Ranges in elevated (> 10 µg/L) maxima of arsenic groundwater concentrations for districts in Bihar

District	**As (µg/L)**
Begusarai	10.0 – 50.0
Bhagalpur	10.0 – 30.0
Bhojpur	10.0 – 50.0
Buxar	20.0 – 40.0
Dhanbad	57.0
Dharbhanga	10.0
E. Champaran	10.0 – 40.0
Godda	60.0
Gopalganj	10.0
Katihar	10.0 – 40.0
Khagaria	10.0 – 40.0
Lakhisarai	20.0 – 50.0
Lohardaga	10.0
Madhepura	20.0
Muzaffarpur	40.0
Purnea	10.0 – 20.0
Saharsa	10.0 – 50.0
Samastipur	10.0 – 40.0
Siwan	20.0
Vaishali	10.0 – 40.0
W. Champaran	10.0 – 20.0

C3: Ranges in elevated (> 10 µg/L) maxima of arsenic groundwater concentrations for districts in Uttar Pradesh

District	As (µg/L)
Azamgarh	10.0 - 811
Badaun	30.0
Bahraich	10.0 – 55.0
Basti	10.0 – 20.0
Deoria	10.0 – 86.0
Gorakhpur	10.0 – 40.0
Jhansi	20.0
Kausambi	20.0
Kushinagar	10.0 – 30.0
Lakhimpur	63.0
Maunath Bhanjanm	50.0 – 82.0
Pilibhit	10.0
Shahjahanpur	10.0 – 20.0

C4: Mean value and range in arsenic groundwater concentrations for two locations in the Haridwar district (Uttarakhand)

Location	As (µg/L)
Bhagwanpur block	12.5 (mean value)
Laksar block	0 – 84.0

C5: Ranges in maxima of arsenic groundwater concentrations for states in Northeastern India

District	State	As (µg/L)
Cachar	Assam	65.0
Barpeta	Assam	100 – 200
Darrang	Assam	200
Dhemaji	Assam	100 – 200
Dhubri	Assam	100 – 200
Golaghat	Assam	10.0 – 200
Jorhat	Assam	10.0 - 657
Lakhimpur	Assam	20.0 – 550
Nagaon	Assam	10.0 – 112
Nalbari	Assam	20.0 – 422
Sibsagar	Assam	20.0
Sonitpur	Assam	10.0
Thoubal	Manipur	798 – 986
Papum Pare	Arunachal Pradesh	74
West Kameng	Arunachal Pradesh	127
East Kameng	Arunachal Pradesh	58
Lower Subansiri	Arunachal Pradesh	63 – 159
Dibang Valley	Arunachal Pradesh	75 – 618
Tirap	Arunachal Pradesh	90
West Tripura	Tripura	191
Dhalai	Tripura	65 – 444
North Tripura	Tripura	122 - 283
Mokok Chong	Nagaland	50 – 278
Mon	Nagaland	67 - 159

C6: Averages of arsenic groundwater concentrations < 10 µg/L in Uttarakhand, Sikkim and Mizoram

District	State	As (µg/L)
Dehradun	Uttarakhand	1.00
Haridwar	Uttarakhand	0.67
Pauri	Uttarakhand	0.33
Tehri	Uttarakhand	1.00
Chamoli	Uttarakhand	0.50
Uttarkashi	Uttarakhand	0.33
Rudraprayag	Uttarakhand	2.50
Nainital	Uttarakhand	0.33
Almora	Uttarakhand	0.17
Pithoragarh	Uttarakhand	0.17
Bageshwar	Uttarakhand	1.00
Champawat	Uttarakhand	0.67
Udham Singh Nagar	Uttarakhand	0.67
-	Sikkim (no specific location indicated in literature)	< 2.0
-	Mizoram (no specific location indicated in literature)	< 10.0

Appendix D: Extended data on surface geology

D1: Surface geology of arsenic affected districts in West Bengal

District	Surface geology	Sediments and rock characteristics
Burdwan, Hooghly, Malda, Murhsidabad	Newer alluvium + Older alluvium	Silt, clay, sand and peat
Howrah, Kochbihar, Nadia, North 24 Parganas, South 24 Parganas	Newer alluvium	Silt, clay, sand and peat

D2: Surface geology of arsenic affected districts in Bihar

District	Surface geology	Sediments and rock characteristics
Begusarai, Bhagalpur, Bhojpur, Buxar, Dhanbad, Dharbhanga, E. Champaran, Godda, Gopalganj, Katihar, Khagaria, Lakhisarai, Lohardaga, Madhepura, Muzaffarpur, Purnea, Saharsa, Samastipur, Siwan, Vaishali	Newer alluvium	Gravel, sand, clay, silt and pebble
W. Champaran	Newer alluvium + Older alluvium Vindhyan Supergroup	Sand, silt, clay, pebble, cobbles, pebble and gravel. Sandstone, limestone and dolomite

D3: Surface geology of arsenic affected districts in Uttar Pradesh

District	Surface geology	Sediments and rock characteristics
Azamgarh, Badaun, Bahraichm, Basti, Deoria, Gorakhpur, Kausambi, Kushinagar, Lakhimpur, Maunath Bhanjanm, Pilibhit, Shahjahanpur	Newer alluvium	Sand (locally micaceous), silt and clay
Jhansi	Bundelkhand Granitoid Complex	Porphyritic granite and leucogranite

D4: Surface geology of arsenic affected districts in North-eastern India

District	State	Surface geology	Sediments and rock characteristics
Cachar	Assam	Barail Group	Siltstone, shale, coal seam and sandstone
Barpeta, Darrang, Dhemaji, Dhubri, Golaghat, Jorhat, Lakhimpur, Nalbari, Sibsagar, Sonitpur	Assam	Fluvial sediments	Sand, silt and clay
Nagaon	Assam	Fluvial sediments, Shillong Group	Sand, silt and clay. Quartzite, phyllite and schist
Thoubal	Manipur	Disang Group	Ophiolites
Papum Pare, East Kameng, Lower Subansiri	Arunachal Pradesh	Bomdila Group	Schist, phyllite, metavolcanics and quartzite
West Kameng	Arunachal Pradesh	Dirang Formation, Seta Group	Phyllite, quartzite and mica schist. Gneiss and high grade schist
Dibang Valley	Arunachal Pradesh	Bomdila Group, Lohit Granitoid Complex	Schist, phyllite, metavolcanics and quartzite. Granite, diorite, granodiorite, leucogranite and tonalite.
Tirap	Arunachal Pradesh	Disang Group	Sandstone and shale
West Tripura, Dhalai, North Tripura	Tripura	Dupi Tila Formation, Tipam Formation, Shurma Group	Sandstone with conglomerate and shale. Siltstone, shale, clay and conglomerate. Siltstone, mudstone, sandstones and shales
Mokok Chong, Mon	Nagaland	Barail Group, Disang Group	Siltstone, shale, coal seam and sandstone. Sandstone and shale