

**Effect of land use and land cover change on groundwater  
Fluoride and Arsenic concentration and associated health  
risks**



by

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PAKISTAN**

**2023**

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This work is submitted in partial fulfilment for the award of the degree of

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**in**

**Environmental Sciences**



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**2023**

## **Certificate of Approval**

It is to certify that the research work presented in this Thesis, entitled “**Effect of land use and land cover change on groundwater Fluoride and Arsenic concentration and associated health risks**” was conducted by **Tahira Nadir (Reg. No. 02312113011)** under the supervision of **Dr. Abida Farooqi**. No part of this thesis has been submitted else for any other degree. This thesis is submitted to the **Department of Environmental Sciences**, in partial fulfilments of the requirements for the degree of **Master of Philosophy** in the field of Environmental Science, Quaid-i-Azam University, Islamabad, Pakistan.

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I **“Tahira Nadir (Reg No. 02312113011)”** hereby state that my M.Phil. thesis titled **“Effect of land use and land cover change on groundwater Fluoride and Arsenic concentration and associated health risks”** was carried out by me in the Hydro-geochemistry Laboratory, Department of Environmental Sciences, Quaid-i-Azam University, Islamabad. The results, findings, conclusions, and investigations of this research has not been previously presented and published as research work in any other university.

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## List of Figures

<b>Figure 1:</b> Global total water distribution.....	9
<b>Figure 2:</b> Sources of groundwater contamination.....	10
<b>Figure 3:</b> Sources of groundwater Fluoride .....	12
<b>Figure 4:</b> Arsenic cycle and its sources in groundwater .....	14
<b>Figure 5:</b> Spatial distribution of Nitrate contamination in (a) 1993 and (b) 2014 in Northern Kelantan .....	18
<b>Figure 6:</b> Study Area, Lahore.....	22
<b>Figure 7:</b> Methodology flow chart. ....	23
<b>Figure 8:</b> LULC maps, A 1995, B 2006, C 2011, D 2016, E 2021 .....	31
<b>Figure 9:</b> Areal coverage of different LULC in different years.....	33
<b>Figure 10:</b> Fluoride mean in different LULC. ....	37
<b>Figure 11:</b> Arsenic mean in different LULC. ....	38
<b>Figure 12:</b> Fluoride geo-statistical results, A represents 2006, B represents 2011, C represents 2016, D represents 2021. ....	41
<b>Figure 13:</b> Arsenic geo-statistical results, A represents 2006, B represents 2011, represents 2016 and D represents 2021.....	44

## List of Tables

<b>Table 1:</b> Worldwide Fluoride and Arsenic co-occurrence in groundwater.....	16
<b>Table 2:</b> Effect of LULC on groundwater quality.....	20
<b>Table 3:</b> Remote sensing data .....	22
<b>Table 4:</b> Accuracy assessment of the classified maps for the year 1995, 2006, 2011, 2016 and 2021.....	<b>Error! Bookmark not defined.</b>
<b>Table 5:</b> Net change (in %age) in LULC from the year 2006 to 2021. ....	34
<b>Table 6:</b> Fluoride data for groundwater in different years, in the respective LULC classes. .	35
<b>Table 7:</b> Arsenic data of groundwater in different years, in the respective LULC classes.....	36
<b>Table 8:</b> Zonal statistical table for Fluoride. ....	40
<b>Table 9:</b> Zonal statistical table for Arsenic. ....	43
<b>Table 10:</b> Human health risk assessment for Fluoride. ....	47
<b>Table 11:</b> Human health risk assessment for Arsenic. ....	48

## Table of Content

Certificate of Approval .....	iii
Author's Declaration.....	iv
Plagiarism Undertaking .....	v
Acknowledgment .....	vi
List of Figures .....	vii
List of Tables .....	vii
Table of Content .....	viii
Abstract.....	2
1. INTRODUCTION .....	3
1.1 Concept of Land Use and Land Cover Changes (LULC) .....	3
1.2 Background .....	3
1.3 Literature Review on LULC .....	4
1.4 Causes of LULC Changes .....	6
1.5 LULC and Groundwater.....	8
1.5.1 Groundwater Contamination .....	8
1.5.2 Fluoride.....	11
1.5.3 Arsenic.....	13
1.6 Co-occurrence of Arsenic and Fluoride in Groundwater .....	14
1.7 Fluoride and Arsenic occurrence in Pakistan .....	16
1.8. Impacts of LULC on Groundwater Quality .....	17
1.9. Impacts of LULC on Groundwater Fluoride and Arsenic.....	19
1.10 Problem Statement .....	21
1.11 Objectives.....	21
2. MATERIALS AND METHOD.....	21
2.1. Study Area.....	21
2.2.1. Climate and Geology of the Study Area.....	21
2.2. Data Sets and Methods.....	22
2.2.1. Data Acquisition .....	22
2.2.2 Methods and Data Processing.....	23
2.2.3 Data Processing .....	23
2.2.4 Atmospheric and Radiometric Correction.....	24
2.3. Land Use Land Cover (LULC) Classification and Accuracy Assessment .....	24
2.4. Groundwater Fluoride and Arsenic Concentration Data.....	25
2.5 Relation between groundwater Fluoride, Arsenic and LULC.....	26



2.5.2 Classification of Groundwater Fluoride and Arsenic Concentration .....	28
2.6. Human Health Risk Assessment .....	28
2.6.1 Exposure Assessment .....	28
2.6.2 Human Health Risk Assessment.....	<b>Error! Bookmark not defined.</b>
3. RESULTS AND DISCUSSION .....	30
3.1 Accuracy Assessment of LULC .....	30
3.2 Land Use and Land Cover Changes .....	30
3.2.1. Land use land cover change 1995.....	30
3.2.2. Land use land cover change 2006.....	30
3.2.3 Land Use Land Cover Change in 2011 .....	31
3.2.4. Land Use Land Cover Change in 2016 .....	32
3.2.5 Land Use Land Cover Change in 2021 .....	32
3.3 Change in Land Use Land Cover .....	34
3.4 Groundwater Fluoride and Arsenic Concentration .....	35
3.5 Groundwater Fluoride and Arsenic Concentration in Different Land Use Land Cover Classes.....	37
3.5.1 Fluoride.....	37
3.5.2 Arsenic.....	38
3.6 Relation between groundwater Fluoride, Arsenic concentration, and land use land and land cover change.....	39
3.6.1. Fluoride.....	39
3.7.2. Arsenic.....	43
3.8. Human Health Risk Assessment .....	47
3.8.1. Fluoride.....	47
3.8.2. Arsenic.....	48
4. Conclusion .....	50
5. References.....	51

## Abstract

Groundwater contamination has become an issue of concern worldwide. A huge amount of population is living in cities and urbanized areas and are highly vulnerable to toxic contaminants. One major cause of groundwater contamination is land use and land cover changes. This has a considerable impact on the groundwater quality and ultimately on the human health. Lahore, a highly urbanized city of Pakistan, relies heavily on groundwater for drinking purpose which is polluted with harmful contaminants. This study examines the spatial and temporal variations in the groundwater Fluoride and Arsenic concentration in Lahore city for the years 2006, 2011, 2016 and 2021. The relationship between variation in groundwater Fluoride and Arsenic concentration and land use and land cover changes is also determined. The associated health risk is also assessed using carcinogenic and non-carcinogenic risk assessment. The results for spatial changes showed a remarkable increase in the built up class from 11.5% in 1995 to 28.6% in 2021 with a net change of +17.1%. While vegetation class decreased from 81.3% in 1995 to 54.4% in 2021 with a net change of -26.95%. Arsenic concentration showed a distinct increasing pattern in the built-up class being 118.3 $\mu\text{g/l}$  in 2006 to 145.223 $\mu\text{g/l}$  in 2021 followed by barren land which increased from 106.66 in 2006 to 141.70 in 2021. While Fluoride showed less distinct pattern but was observed to be increasing with the passage of time. This consequently increased the health risk in the residents with Fluoride non-carcinogenic risk increasing from 3.01 in 2006 to 4.51 in 2021 while carcinogenic risk for Arsenic increased from 0.0041 in 2006 to 0.0067 in 2021. This study gives an insight into the association between land use and land cover changes and ground water quality and also provides suggestions for the policy makers to design reasonable land use and land cover strategies to avoid groundwater contamination and hence protect the health of the residents.

## 1. INTRODUCTION

### 1.1 Concept of land use and land cover change (LULCC)

Land use and land cover changes refer to changes that are made in the pattern of land use and land cover and the human-induced modifications in the natural pattern of land cover. (Elbeih & El-Zeiny, 2018). Land use and land cover have been defined as distinct concepts with different meanings. Land cover refers to the physical characteristics of the Earth's surface, including water bodies, soil, vegetation, and hard surfaces such as mountains. On the other hand, land use refers to the ways in which people utilize or exploit the land for various purposes such as settlements, agriculture, forestry, and pasture, which can disrupt natural processes such as biogeochemistry, hydrology, and biodiversity. In recent decades, activities by humans such as deforestation, agriculture and urbanization have significantly changed the Earth's surface. To maintain or modify a specific land cover type, people undertake various activities, inputs, and arrangements, which is the definition of land use according to Abebe (2018). Areas that were forests and grasslands are now being converted to urban lands and industrial lands which reduces the green portion of the land and the excessive industrialization and rapid urbanization results in environmental degradation.

### 1.2 Background

The study of the distribution and history of land use and land cover (LULC) has been recognized as essential over time. Agricultural practices and timber harvesting were significant causes of the partial clearing of large areas of Western Europe by 1500. The impact of LULCC (Land Use and Land Cover Change) was particularly strong in Western Europe. Furthermore, significant LULCC occurred in most of Asia, including India and China, by 1800. As early as 1750, approximately 6-7% (7.9-920 million square kilometers) of the Earth's land surface was under cultivation (Goldewijk, 2001).

According to Goldewijk (2001), the primary drivers of Land Use and Land Cover Change (LULCC) worldwide are the intensification and expansion of agriculture. It has been estimated that the global arable land area has raised from 300 million hectares in 1700 to 53 million hectares in 2000. However, there are discrepancies among estimates from various sources. Ramankutty and Foley (1999) calculated 4 million hectares in 1700 and 20 million hectares in 1990, while Savage and Williams (1990) estimated 14 million

hectares and Richards (1990) estimated 15 million hectares, although both of these estimates were for the year 1980. Matthews (1983) estimated a higher figure of 17.6 million hectares of farmland in 1980. The current estimate for under-irrigated agricultural land ranges from 250 million to 274 million hectares, as reported by Siebert et al. (2005). The expansion of arable land worldwide has been remarkable, currently accounting for nearly 11% of the total land area (Biemans et al., 2011). Similarly, grazing livestock areas have increased rapidly, from 324 million hectares in 1700 to 3429 million hectares in 2000, which is approximately 25% of the overall area of land (Richards, 1990). Urban areas, on the other hand, constituted less than 1% of the total land area until the mid-20th century, and even now, they make up less than 5% of the global total land area (Potere & Schneider, 2007).

### **1.3 Literature review on land use and land cover change**

The migration of people to urban areas every year results in swift transformations in the ecosystem, natural landscapes, topography, and environment (Hokao & Phonekeo, 2012). Such changes in the topography of the urban areas involve deforestation, reduction in agricultural land due to the expansion of settlements, and the increase of barren land (Kumar et al., 2012). Landscapes are subject to change due to natural causes such as floods, earthquakes, and erosion, as well as anthropogenic causes such as urbanization, deforestation, and climate change. However, significant changes have occurred in recent decades, primarily due to the rapid growth of cities (Kumar et al., 2012).

It is estimated that nearly 3.5 billion individuals currently reside in urban areas, and this figure is expected to further rise to comprise 60% of the total global population by 2030. Furthermore, by 2025, the number of large cities is projected to reach 100 (Avelar et al., 2009). The increasing trend of urbanization worldwide has stimulated numerous researchers to investigate the potential impacts of human induced activities on the environment, including the quality of groundwater. The primary drivers of significant Land Use Land Cover Change are the transformation of natural habitats for human habitation and the expansion of agriculture to support the increasing population.

Land Use and Land Cover (LULC) have been subject to significant changes due to human activities in both developed and developing countries over the course of centuries (Liu & Tian, 2010). The analysis of LULC has become increasingly important in recent decades,

as it plays a crucial role in reducing biodiversity, altering ecosystems, and modifying micro and macro climate patterns and compositions (Aguilar et al., 2003). Land Use and Land Cover (LULC) changes caused by human activity have had significant impacts on terrestrial ecosystems at local (Campra et al., 2008), regional (Hu et al., 2010), and global (Lawrence et al., 2012) scales, affecting the environment in various ways (Guo et al., 2012). In the past few decades, the expansion of built-up areas, including buildings and concrete/asphalt surfaces, has increased from 54% in 1975 to 61% in 2000, while there has been a significant loss of vegetation cover, reducing from 38% in 1975 to 33% in 2000 in Merseyside, England, by 6% (Pauleit et al., 2005). The complex interaction between LULC change and environmental factors impacts human populations in different ways. The detection and mapping of LULC changes are crucial for various disciplines, such as urban planning and management, climate change, and environmental monitoring (Senf et al., 2015).

The Adama Zuria District in Ethiopia experienced a significant increase in settlement area from 0.73% in 1989 to 2.28% in 1999 and 6.11% in 2016, accompanied by a growth in plantation area from 7.41% in 1989 to 7.57% in 1999 and 8% in 2016. The proportion of shrub-land decreased from 21% in 1989 to 12.50% in 1999, and further down to 12.43% in 2016. Agricultural land, on the other hand, increased from 61.7% to 69.15%, while bare land decreased from 8.89% in 1989 to 3.98% in 2016 (Tafesse & Suryabhadgavan, 2019). These changes in land cover and pattern have been identified as the primary drivers of environmental degradation including groundwater degradation, alterations in urban hydrology, rising urban thermal islands, and climate change at local to regional scales (Zhou et al., 2016).

In Pakistan, like many other countries, the issues of fast population growth, urbanization, and changes in land use and cover have become a major concern. Previous research indicates significant transformations in land use. For instance, in the Kalam region, the forest area has decreased by 30.5%, with 11.4% resulting from deforestation caused by agricultural expansion. Moreover, agricultural land has decreased by 17.3% and has been converted into pasture (Qasim et al., 2011). Another study revealed that LULC has led to the loss of vegetation cover and a reduction in open areas and agricultural land, both inside and outside the city. The research further confirmed that the growth of the city has

mainly occurred in the agricultural land that used to be part of rural area. Urban sprawl has increased considerably in the district since 1973 (Shirazi & Kazmi, 2014).

#### **1.4 Causes of land use and land cover change**

Land use and land cover changes provide insights into the past and potential future of human civilization. These changes are influenced by various factors, including population growth, economic development, technological advancements, and environmental transformations (Houghton, 1994). Among these factors, population growth stands out as a major driver of land use and land cover change. People are both independent entities and interconnected with each other, crucial for sustainable development, as humans represent the most valuable natural resource (Abebe, 2018). Over the past three centuries, the total cultivated area on Earth has expanded significantly, by approximately 45% or more, increasing from 2.65 million km<sup>2</sup> to 15 million km<sup>2</sup>. This expansion has primarily been driven by agriculture and the expansion of urban areas. However, this growth has come at the expense of other natural resources such as forests, which have dwindled due to land conversion and urbanization.

Land use and land cover changes are recognized as significant global issues due to their profound impact on environmental dynamics worldwide (FAO, 1999). Various factors contribute to these changes, including deforestation, population growth, urban sprawl, and natural phenomena such as shifts in global systems and cycles. Throughout history, the expansion of agricultural land has been identified as a primary driver of land use and land cover changes (Houghton, 1994).

According to the United States Environmental Protection Agency (USEPA, 2004), the causes of land use and land cover changes can be classified into three general categories:

1. Natural processes: These include atmospheric and climate changes, wildfires, and other natural occurrences that can alter the landscape.
2. Direct effects of anthropogenic activities: Human-driven actions like deforestation and illegal construction directly contribute to changes in land use and land cover.
3. Indirect effects of anthropogenic activities: Activities such as water diversion, which can lead to a decline in the water table, have indirect consequences on land use and land cover patterns.

Urbanization is widely recognized as a significant driver of land use and land cover changes on a global scale. It refers to the ongoing growth in population, density, and residential areas within cities, as well as the expansion of urban areas in terms of their spatial extent (Alig et al., 2004). According to the "New World Encyclopedia," Urbanization can be understood as the increase in the population residing in cities relative to the rural population within a given region. In essence, urbanization entails the process of urban growth and the associated transformation of land use and land cover patterns as rural areas are converted into urban landscapes to accommodate the expanding population and infrastructure needs of cities. Urbanization is commonly understood as a long-term transformation in land use patterns that occurs alongside population and economic growth (A. S. Mather, 1986). One of the prominent forms of urbanization throughout human history is the conversion of land use and land cover. Over time, the urban population has experienced significant growth, reaching 54% globally in 2014 compared to 30% in 1950 (United Nations and Department of Economic and Social Affairs, 2015a). This has led to an expansion of urban areas, often surpassing the rates of population growth (Seto et al., 2011). Projections indicate that this trend of urbanization is likely to continue, with certain regions, such as the Middle East, experiencing even more rapid acceleration (Seto et al., 2012; United Nations and Department of Economic and Social Affairs, 2015b). The patterns of urbanization described above can have significant impacts on various aspects, including the microclimate of urban areas, water quality, and biodiversity, as highlighted by Seto et al. (2012). Urban sprawl leads to the degradation, loss, and transformation of fertile farmland, presenting another important consequence of urbanization.

The patterns of urbanization described above can have significant impacts on various aspects, including the microclimate of urban areas, water quality, and biodiversity, as highlighted by Seto et al. (2012). Urban sprawl, in particular, leads to the degradation, loss, and transformation of fertile farmland, presenting another important consequence of urbanization.

A research investigation conducted in Saharanpur City, India, utilizing GIS and Remote Sensing techniques, focused on examining the impact of urban sprawl and the loss of agricultural land between 1988 and 1998. The study revealed a noteworthy expansion of residential areas attributed to population growth and unplanned urban expansion,

resulting in the irreversible destruction of fertile agricultural land that cannot be reclaimed (Fazal, 2000). The findings from this study highlighted a substantial increase in the urban area of Saharanpur City.

The effects of LULC have been seen in different compartments of the environment. One of these is groundwater, which is a natural resource and currently is in focus due to being contaminated by different activities.

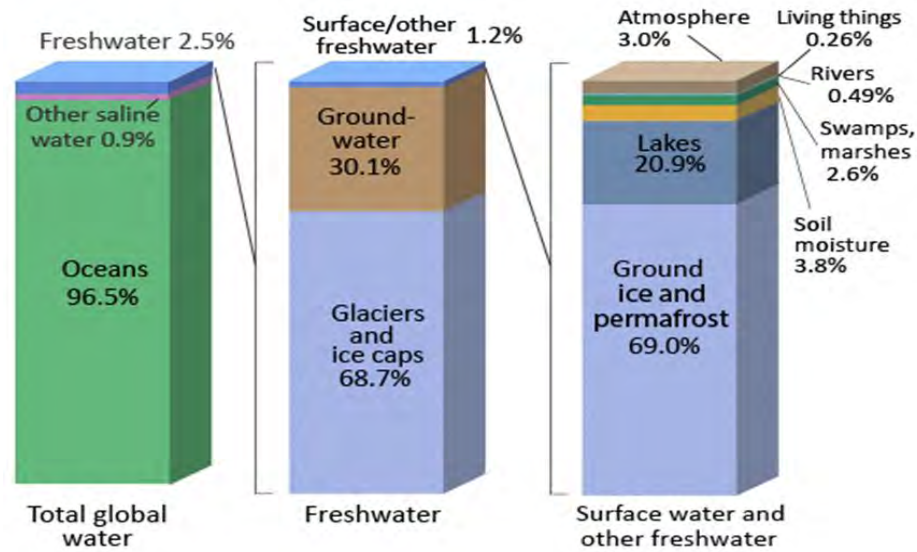
## **1.5 Land use and land cover change and groundwater quality**

### **1.5.1 Groundwater contamination**

Groundwater is a major water resource that plays an important role in sustaining a number of socio-economic activities and maintaining ecosystems and their services (Shakerkhatibi et al., 2019a). Water covers 1386 million km<sup>3</sup> of Earth. Out of 10.63 million km<sup>3</sup> of freshwater, only 30.1% happens as groundwater (Jha & Tripathi, 2021). Considering the water supply of the world, it reveals that saline water in the oceans accounts for 97% of the total and the remaining only 2.5% on the earth's surface is fresh water. Of freshwater, 69% lock up in glaciers and ice caps, the remaining 30% lies below the surface as groundwater and less than 1% is available as an on-ground freshwater source. Lakes hold 20%, soil moisture 4%, rivers 0.49%, and permafrost 70% of total on-ground freshwater (Marshall, S. J. 2013, Gleick, P. H. 1993).

Groundwater acts as a primary water source in arid and semi-arid regions of the world. It is estimated that around 1.5 billion people around the globe depend on groundwater for their drinking requirements (Mukherjee and Singh, 2018). Groundwater is often regarded as the most favorable alternative water source compared to rivers, streams, lakes, dams, etc. because it benefits from natural protection against contamination, unlike surface water. As a result, it undergoes inherent filtration and purification as it percolates during rainy days, reducing the need for extensive treatment processes, including disinfection. (Dehghani et al., 2019).

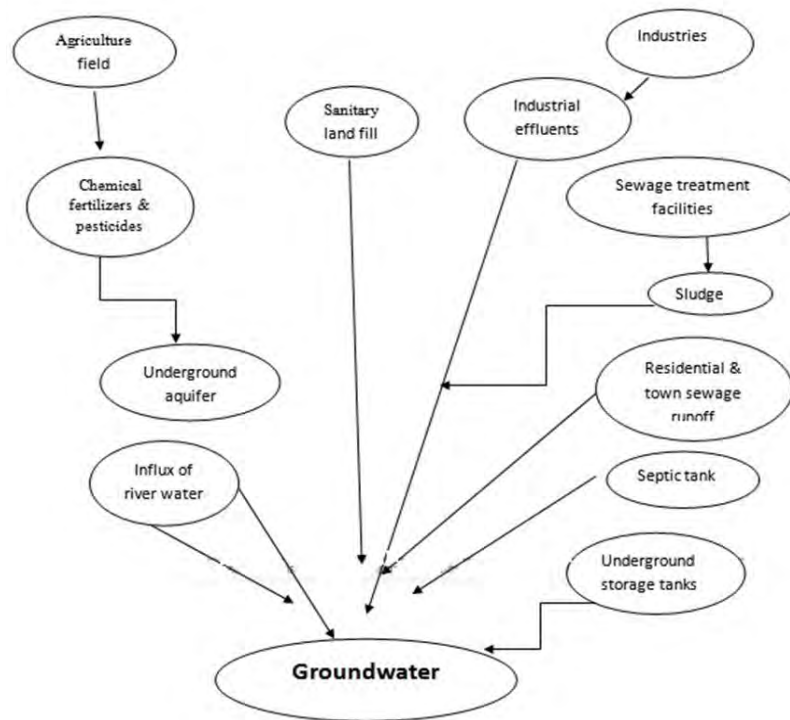




**Figure 1:** Global total water distribution (Gleick, P. H. 1993, Kundzewicz et al., 2007)

Even though groundwater is clearly safe to consume, its quality can be impacted by a few factors, including the aquifer's geology and chemical makeup, the climate, and human activities. But globally, groundwater pollution is becoming an increasingly serious issue, particularly in areas with severe water shortages (Kouadra & Demdoum, 2020). Groundwater has deteriorated and contaminated in different regions (Dosskey & Qiu, 2011). Due to unsustainable use and excessive groundwater pollution, it is reported that around 1.8 billion people may encounter the issue of water scarcity in the near future, by 2025 (Hasana et al., 2019). The quality of the water we drink has a direct impact on our quality of life. According to the WHO report of 2011 (WHO, 2011 Report on Groundwater), nearly 80% of the worldwide diseases and injuries that are leading to death might be due to environmental causes. Contaminated water consumption has resulted in several reported deaths across various regions (P. Li et al., 2021). Obtaining renewable and drinkable groundwater for the people of a country is one of the major contributing factors toward achieving sustainable development. Contamination due to chemicals is considered to be the most threatening issue from the past three decades and has been presented in several groundwater studies (Adimalla et al., 2018). Developing countries are more vulnerable to the effects of low quality groundwater and the residents are exposed to a number of diseases caused by the harmful contaminants. Different contaminants including nanoparticles, microplastic, pesticides, fertilizers and other emerging toxic chemicals like Arsenic and Fluoride are considered as the most

concerning pollutants and have been shown to have caused different human health issues (P. Li & Wu, 2019).



**Figure 2:** Sources of groundwater contamination. Manam, Vishnu Kiran. (2022).

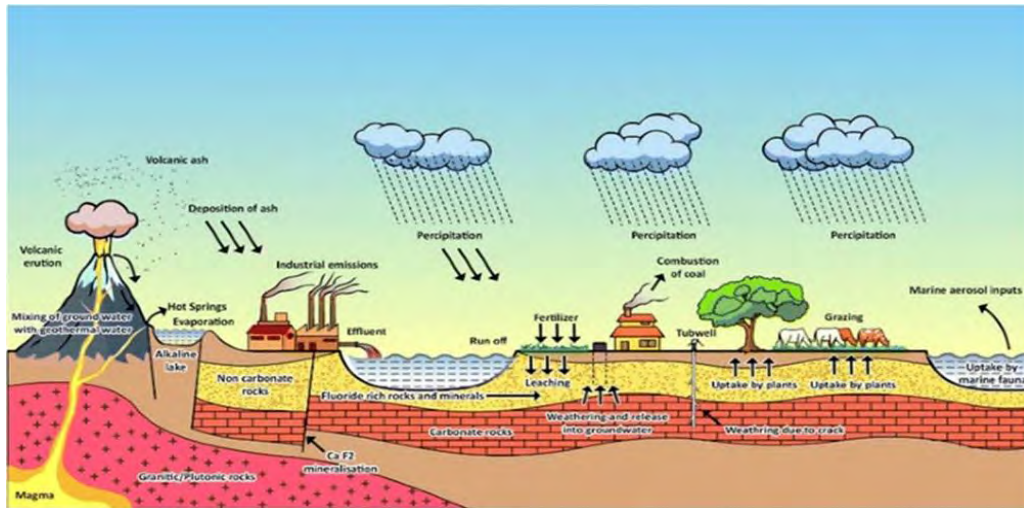
The two most common and emerging contaminants in terms of the number of people affected and the affected area are Arsenic and Fluoride (Sikdar, 2018). In present times, the commonly prevalent inorganic pollutants i.e. Arsenic and Fluoride are in focus owing to their harmful and ubiquitous nuisance (Appelo & Postma, 2005). The contamination of water resulting in a worldwide crisis of water is attributed to the presence of both Arsenic and Fluoride which also cause noteworthy health impacts (Naseem et al., 2010). Generally, As and F enters the groundwater through both geogenic as well as anthropogenic activities. Geogenic causes include rock-water interaction and groundwater recharge while the major anthropogenic sources are rapid urbanization, industrialization, excessive agricultural activities, and other human-induced processes (Nath, 2018). Arsenic and fluoride enter the body through drinking water. Children are more vulnerable to the adverse impacts of Arsenic and Fluoride contaminated water as compared to adults as it has a major effect on their growth as well as intelligence level (Ghosh & Mukhopadhyay, 2018).

## 1.5.2 Fluoride

Fluorine, also referred to as fluoride (F<sup>-</sup>) in aqueous environments, constitutes approximately 0.08% of the earth's crust, ranking as the 13th most abundant element in this geological layer. The periodic table's lightest and most electronegative element is Fluorine (F), which is a member of the halogen group (Narsimha & Sudarshan, 2017). The amount of Fluoride in the Earth's bedrock is high (625 mg/kg), and it is present in water as the negatively charged Fluoride ion (F<sup>-</sup>) (Ali et al., 2016). Fluoride concentrations in seawater, waterways, and lakes are typically 1 mg/L and less than 0.5 mg/L, respectively. In groundwater, its concentration typically lies in the range of 0.1 and 10 mg/L (Dehghani et al., 2019). Fluoride, a naturally occurring element, is widely recognized as one of the most toxic inorganic pollutants found in water.

### 1.5.2.1 Sources of Fluoride

Fluoride levels in water can be high because of both natural processes (weathering of Fluoride-containing rocks and minerals) and human activities (industries like aluminum and zinc smelters, fertilizer and herbicide manufacturing, and electronic manufacturing) (Dehghani et al., 2019). The majority of Fluorine in water comes from geogenic sources. Minerals such as fluorspar, mica, rock phosphate, apatite, fluorite, cryolite, and topaz contain fluoride as an essential component (Banerjee, 2015). Igneous rocks, mineralized veins, and sedimentary formations contain fluorine minerals in abundance (Ali et al., 2016). These minerals' physical, chemical, and biological weathering releases fluoride into soils, which then enters the water (Dehghani et al., 2019). The primary way to consume naturally occurring fluoride is through drinking water. All naturally occurring waterways contain some fluoride.



**Figure 3:** Sources of groundwater Fluoride (Mukherjee and Singh, 2018).

### 1.5.2.2 Impacts of Fluoride exposure

Fluoride is a crucial element for human health, but excessive levels can have severe health consequences. Fluoride permissible limit in drinking water is between 0.5 to 1.5 mg/L, according to world health organization (WHO) guidelines (Dehghani et al., 2019). Various factors contribute to the contamination of groundwater with Fluoride, including pH-dependent dissolution, aqueous ionic concentrations, rock-water interactions, atmospheric deposition, and mobilization through carbonate and bicarbonate ions. More than 200 million people across 25 countries suffer from fluorosis, a condition caused by consuming fluoride-contaminated groundwater (M. Yadav et al., 2021). Fluoride can negatively impact bones, teeth, skeletal muscles, and the nervous system, and its toxicity can lead to symptoms such as salivation, nausea, diarrhea, and abdominal pain (K. K. Yadav et al., 2019). Furthermore, the high uptake of Fluoride has resulted in various socio-economic problems, including the abandonment of boreholes due to groundwater contamination (Jha & Tripathi, 2021).

Despite the fact that Fluoride is found in food, air, and water, drinking water remains the main source of exposure (Vithanage & Bhattacharya, 2015). 65% of endemic fluorosis worldwide is caused by Fluoride contamination in groundwater (Maghanga et al., 2022). Fluoride excessive concentration in groundwater and its threatening impacts on the residents have been reported in different countries like China (X. He et al., 2020; Wen et al., 2013a), Mexico (P. Li et al., 2021), Iran (Yousefi et al., 2019), India (I. Mukherjee et al., 2019) and Fiji (Prasad et al., 2018).

### 1.5.3 Arsenic

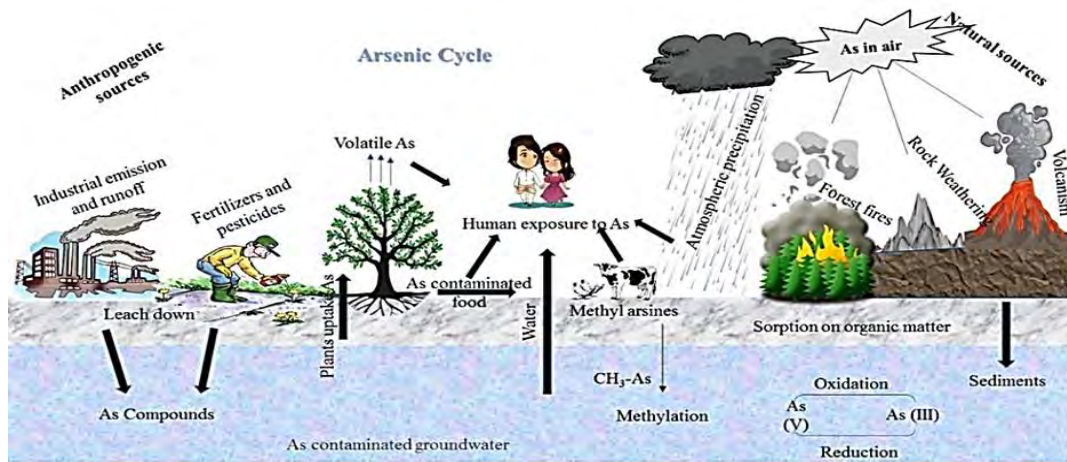
The 33rd element on the periodic chart is Arsenic (As). 1.5 to 2 ppm of elemental Arsenic can be found in the bedrock. Arsenic is a chemical element that has a grayish appearance. It typically coexists with oxygen, sulfur, and chlorine (Shaji et al., 2021). Due to the presence of minerals like arsenopyrite ( $\text{FeAsS}$ ), realgar ( $\text{As}_2\text{S}_2$ ), orpiment ( $\text{As}_2\text{S}_3$ ), and iron pyrites ( $\text{FeS}_2$ ), Arsenic is more frequently detected in groundwater (Nikolopoulos, D.P.D., 2015). Different sorts of water have varying Arsenic concentrations. It fluctuates between 0.15 and 0.45 g/L in freshwater and 1.0 to 3.0 g/L in saltwater (Palit et al., 2019). Arsenic is most often found in the oxidation states of -3 (arsenides), +3 (arsenites), and +5 (arsenates). While As(III) is dominant in a reducing environment, As(V) predominates in an oxidative environment (Zhou et al., 2017). Arsenates also referred to as salts of Arsenic (V) acid, are significant pollutants in groundwater. Arsenic contamination of groundwater is a global issue (Poonia et al., 2021)

While Arsenate (As(V)) is the main form of arsenic found in drinking water, substantial concentrations of the more toxic form Arsenite (As(III)) have also been observed in reducing environments. There are additional types that can exist, including methylated arsenic and organic arsenic (Jiang et al., 2019). The most widespread form of Arsenic in drinking water is organic Arsenic (arsenate plus arsenite). Arsenic concentrations in drinking water that are above WHO limits threaten an estimated 200 million people worldwide (Gan et al., 2014).

#### 1.5.3.1 Sources of Arsenic

Arsenic-enriched rocks are the main source of Arsenic in nature. Volcanoes and eroded arsenic-bearing minerals like arsenopyrite ( $\text{FeAsS}$ ), lollingite ( $\text{FeAs}_2$ ), orpiment ( $\text{As}_2\text{S}_3$ ), and realgar are typical examples of natural sources. (AsS) (Fendorf et al., 2010). Alkali desorption, iron-containing mineral reductive dissolution, sulfide mineral oxidation, crustal processes, geothermal activity, leaching and weathering of silicate and carbonate minerals, redox processes, exothermic processes, and exothermic reactions are the main causes of the excessive groundwater arsenic concentration, according to (A. Mukherjee et al., 2011; Pal et al., 2009). On the other hand, frequent anthropogenic sources include industrial manufacturing, agriculture, and raising livestock. Additionally,

it is extensively utilized in the production of medicines, copper, lead alloys, glassware, industrial chemicals, and copper (Ullah et al., 2023).



**Figure 4:** Arsenic cycle and its sources in groundwater (Masuda, 2018).

### 1.5.3.2 Impacts of Arsenic exposure

Exposure to even low or moderate Arsenic levels ( $10\text{--}300\ \mu\text{g L}^{-1}$ ) in drinking groundwater can have serious human health consequences like skin irritation and lesions, nervous disorders, and liver issues and exposure for longer periods of time can lead to gene mutation and cancers of different types and ultimately death occurs (Uppal et al., 2019). Therefore, WHO has set the permissible level of Arsenic in drinking water to  $10\ \mu\text{g L}^{-1}$  (WHO, 2011). In developing countries, groundwater is consumed without any treatment and it is reported that around 45% of the population in Bangladesh is exposed to the severe effects of groundwater Arsenic which is above the set limit of  $10\ \mu\text{g L}^{-1}$ , which is causing about 24,000 deaths annually (Neumann et al., 2010). Similarly, in China, 20 million people are exposed to the harmful effects of groundwater Arsenic (Hao et al., 2018).

### 1.6 Co-occurrence of Arsenic and Fluoride in groundwater

Co-occurrence in the case of Fluoride and Arsenic is a general term that does not always indicate a favorable relationship between the two contaminants. There were frequently high concentrations of both at the same moment despite the fact that different pathways of release frequently resulted in little or no correlation between the two. Seasonality, environment, and climate are the main determinants of the co-occurrence strength. The

primary cause of the co-occurrence is still anthropogenic forcing, which includes mining, irrigation return flow, extraction, recharge, and agrochemicals (Kumar et al., 2020a). A study was conducted in Mailsi, Punjab by (Rasool et al., 2016), and reported a number of correlations that were very low and not significant at all. While on the contrary, a study conducted in Kalanwala, Punjab indicated the co-occurrence of Fluoride and Arsenic and showed a positive correlation, since both contaminants were found to have few common sources including anthropogenic activities like coal combustion (Farooqi et al., 2007).

However, according to accounts of a positive correlation, the existence of Fluoride and Arsenic in samples of groundwater points to a shared source or pathway of contamination for both elements (Rasool et al., 2016). The primary cause of Fluoride and Arsenic pollution is thought to be water-rock interaction, which may be further hastened by geothermal and mining operations as well as overexploitation of aquifers and many other anthropogenic activities like rapid urbanization, intensive industrialization, and unsustainable irrigation and agricultural activities (Kumar et al., 2020a). Multi-element enrichment of nutrients in groundwater systems is an emerging concern. Their co-occurrences can lead to an enhanced effect through various complex synergistic-antagonistic interactions (Patel et al., 2019). Co-occurrence of As-F endure, according to (Kumar et al., 2016) a number of interconnected, complicated relationships with the groundwater and compared to their separate effects, can have serious negative effects on health. Numerous countries around the world have noticed and documented the coexistence of both contaminants.

**Table 1:** Worldwide Fluoride and Arsenic co-occurrence in groundwater.

Country	Co-occurrence	Reference
<b>Pakistan</b>	A weak correlation was found between As and F. Positive correlation with inorganic Arsenic $r = 0.64$ . Co-occurrence	(Farooqi et al., 2007; Rasool et al., 2016); (Brahman et al., 2013); (Farooqi et al., 2007)
<b>China</b>	No correlation was studied but high concentration and co-occurrence of As and F in groundwater. Significant correlation. Positive correlation due to high soda environment.	(Bian et al., 2012; J. Li et al., 2012; C. H. Wang et al., 2007; Zhang et al., 2003); (Wen et al., 2013b); (Y. Wang et al., 2009)
<b>India</b>	No correlation was studied but a high concentration of As and F. The strength of the correlation was found to be not significant.	(Borah et al., 2010); (Kumar et al., 2016; Patel et al., 2019)
<b>Mexico</b>	Positive correlation between As and F, $r = 0.93$ . Positive correlation in a semi-arid region.	(Armienta & Segovia, 2008); (Reyes-Gómez et al., 2013); Herrera et al., 2017)
<b>Africa</b>	The co-existence of As and F is observed.	(Rango et al., 2010)
<b>Argentina</b>	A positive correlation was found between Arsenic and Fluoride. $r = 0.8$	(Gomez et al., 2009)

### 1.7 Fluoride and Arsenic occurrence in Pakistan

Pakistan is one of the most rapidly urbanizing countries in South Asia and with every passing year the rate of urbanization is increasing. It is reported (Masuda, 2018) that the urban population of Pakistan has grown from 17 % in 1951 to 37% in 2010 and 39.7% in 2017, while an annual growth of 2.77% has been recorded. Moreover, the study also says that half of the population of the country will be urbanized in the coming 10 to 15 years.



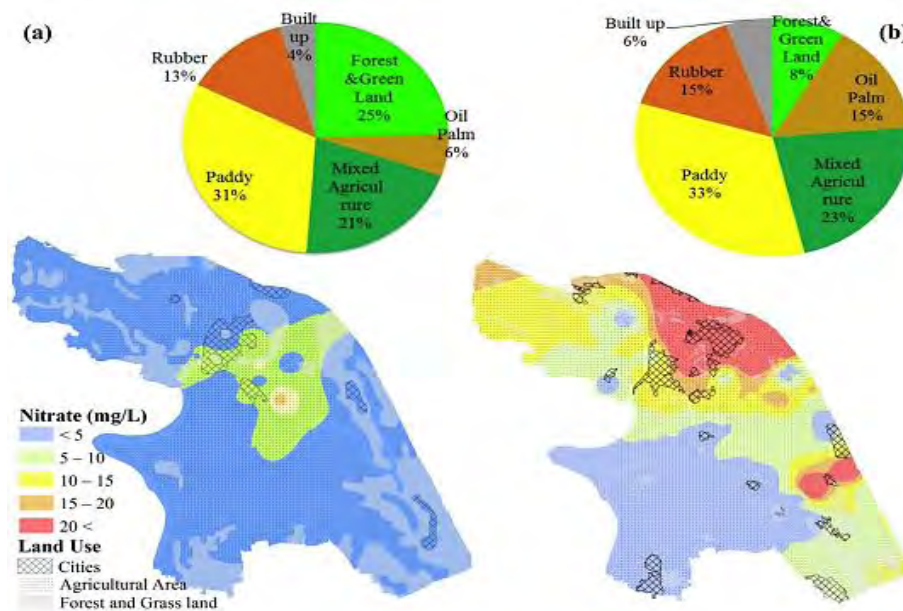
Due to the geological setup, anthropogenic intervention, hydro-geochemistry, and presence of minerals, Pakistan is included among the countries that have high Arsenic and Fluoride contamination in the groundwater (Ali et al., 2016; Masuda, 2018). Several studies have been conducted in Pakistan and they have shown areas where there is high Fluoride toxicity such as Quetta in Balochistan, Kalanwala in Punjab, Dera Ismail Khan in KPK, and Thar in the province of Sindh (Ahmad & Qadir, 2011; Farooqi et al., 2007; Rafique et al., 2009). Similar research reports have been published on the severity of Arsenic issue in Pakistan which highlights that around 25-36% of the people in Punjab and Sindh are vulnerable to Arsenic pollution in drinking water and because of their natural geological origin, the two cities of Punjab, Lahore, and Kasur are considered as the hotspot areas for Arsenic and Fluoride pollution (Rehman et al., 2022).

### **1.8. Impacts of LULC changes on groundwater quality**

The land use changes that are induced by human activities have been shown to affect the groundwater quality in different regions of the world. There have been several studies that show that groundwater quality changes with changes in land use patterns (Bawa & Dwivedi, 2019) and it has been shown that there is a remarkable association between human-induced land use and land considered one of the main alterations in land use and land cover pattern that affects the environment (Mahlknecht et al., 2008). The major cause of the land use and land cover change is to be highlighted and analyzed in order to get a refined solution to reducing groundwater pollution as groundwater reservoirs are more susceptible to the changes made on land and the activities on land surfaces (S. He et al., 2020). In Bangladesh, research was performed to assess the quality of groundwater and to evaluate the impacts of land use changes on groundwater quality. The results indicated an increased level of total dissolved salts in the groundwater which was attributed to the intrusion of seawater into the coastal aquifers as a consequence of excessive agricultural activities (Salman et al., 2018). With every passing day, the population per capita is increasing and the available natural resources are being used in an unmanaged way which is causing the deterioration of the resources. A study was conducted in lower Shivalik hills, Rupnagar Punjab, India which shows that during the time period of 1989 to 2005, a continuous reduction in river area (17%), dense forests (22%), and cropland (31%) was observed while the settlement area increased 534% significantly. These changes ultimately affected the groundwater of the area. The water tasted brackish due to the high

EC value, and a high concentration of nitrates was found in it due to excessive use of fertilizers and leaching. Moreover, high concentrations of different ions like Ca, Mg, Zn, Cd, Cr, and Mn were observed (Singh et al., 2010).

A similar study was conducted in Northern Kelantan Malaysia where the effect of the conversion of grasslands and forests into farmlands and urban lands, on the quality of groundwater is analyzed for the time period of 1989 to 2014. The results indicated a noticeable increase in the concentration of nitrates in the groundwater of the region. Moreover, using the ARIMA model, the study also predicted a 2.6% rise in nitrate contamination and a 3.9% annual increment till 2030 in the residential and agricultural areas (Sheikhy Narany et al., 2017).



**Figure 5:** Spatial distribution of Nitrate contamination in (a) 1993 and (b) 2014 in Northern Kelantan (Sheikhy Narany et al., 2017).

In order to evaluate the linkage between land use and land cover conversions on groundwater quality, research was conducted in Xi'an City in Guanzhong Basin over the years 2005 to 2015. The results of the study indicated that the urban area of the city has expanded up to 160% while the agricultural and forest areas have been reduced by 83.08% and 52.54% respectively. The study concludes that the changes in land use and land cover patterns have shown negative impacts on the groundwater of the area by adding contaminants like  $\text{NO}_3$ , TDS,  $\text{HCO}_3$ , Ca, Mg, and Na. It was shown in the study that

urban land, agricultural land, and industrial lands have shown a negative impact on groundwater while the forests have been seen to have a positive impact on the groundwater (Lee et al., 2021).

### **1.9. Impacts of LULCC on groundwater Fluoride and Arsenic**

Fluoride and Arsenic concentration in groundwater have also changed with the changes in land use and land cover change patterns which have ultimately affected the overall quality of groundwater and thus are known to cause a number of health issues. The effect of land use and land cover changes on the aquifers of arid and semi-arid regions of the world has been studied in different research and the results have shown that due to changes in land use and land cover pattern, different anthropogenic contaminants have been added to the aquifers (Mahlknecht et al., 2008). A study was done in southern Taiyuan city, China where land changes were increasingly seen with rapid industrialization and urbanization in the region. The data collected from 2010 to 2017 showed that in the groundwater of the region, Fluoride concentration ranged between 0.10 and 5.70 mg/l while Arsenic concentration ranged between 0.50 and 71.50  $\mu\text{g/l}$ . These results are attributed to increased land use changes with more of the land being converted to industrial land in the study region (Yuan et al., 2020). Likewise, in Multan, Pakistan, the River Chenab which is the source of groundwater recharge is contaminated with Arsenic due to increased and rapid changes in land use as the solid waste dumping sites increased. When this water recharges the groundwater then, it causes an increased Arsenic concentration in it (Abbas et al., 2022).

A study was done in Northern Mexico, where two aquifers were investigated Aldama-San Diego (ASD) and Tabalaopa-Aldama (TAB) to find the possible association between land use and land cover changes to the aquifer quality. The results showed that the concentration of Arsenic and Fluoride was increased to levels that were higher than the Mexican guidelines for drinking water. Though Arsenic and Fluoride are considered natural contaminants, their toxic concentration in the wells is a consequence of human stress including land use and land cover changes. Specifically, in the study area, a significant loss of grassland and shrub lands observed from 1993 to 2012 (Reyes-Gómez et al., 2013).

**Table 2:** Effect of LULCC on groundwater quality.

Study Area	Effects	Causes	Reference
Bangladesh	Increased level of TDS	- Excessive agricultural activities - Intrusion of seawater into coastal aquifer	(Salman et al., 2018)
Xiaotjiang Watershed, China	Increased pH value, increased $Mg^{2+}$ , $NH_4^+$ , $SO_4^{2-}$ , $NO_3^-$ , $NO_2^-$ , $Cl^-$	Diffusion of excess fertilizers and other pollutants due to the increased cultivated area.	(Jiang et al., 2019)
Lower Shivalik hills, Rupnagar Punjab, India	High EC value, high concentration of nitrates, Ca, Mg, Zn, Cd, Cr, and Mn.	- Continuous reduction in river area (17%), dense forests (22%), cropland (31%) - Settlement area increased	(Singh et al., 2010)
Northern Kelantan Malaysia	Increase in the concentration of nitrates.	Conversion of grasslands and forests into farmlands and urban lands.	(Sheikhy Narany et al., 2017)
Sohag Governorate, Egypt	Increased urbanization	Increased values for TH, EC, TDS, SAR and $SO_4$ , Phosphate, and Silicate levels.	(Elbeih & El-Zeiny, 2018)
South Korea	Excessive use of fertilizers and livestock activities.	$NO_3$ -N increased	(Lee et al., 2021)
Shijiazhuang, Hebei Province, China	- Ground water table reduced - Groundwater shifted to $HCO_3^-$ $SO_4^{2-}$ , Ca (Mg) type from $HCO_3^-$ Ca (Mg) type - High $NO_3^-$ level	Rapid urbanization	(Hao et al., 2018)
Taiyuan city, China	- Increased level of As (0.50 and 71.50 $\mu g/l$ ). - Increased F (0.10 and 5.70 mg/l).	Rapid industrialization and urbanization.	(Yuan et al., 2020)
Multan, Pakistan	Increased Arsenic concentration	Increased solid waste dumping site.	(Abbas et al., 2022)
Northern Mexico	Increased concentration of Nitrates, As, and F.	- loss of grassland and shrubland. - increasing irrigated agricultural use and urban land	(Reyes-Gómez et al., 2013)

### **1.10 Problem Statement**

Lahore is a city with high population and with every passing year, the rate of urbanization and industrialization is increasing in the city. This results in changes in the natural pattern of land which causes obvious alterations in the natural setup of the city. Moreover, Lahore is considered a hotspot area in terms of Arsenic and Fluoride groundwater contamination, which has affected the health of the residents causing serious health issues.

Keeping in view the above scenario, this study investigates whether there is any association between the human-induced alterations in land use and land cover patterns of the city and groundwater quality in terms of Arsenic and Fluoride contamination. If there is any linkage, then how is this going to affect the health of the residents.

### **1.11 Objectives**

This study tends to present the association between changes in land use and land cover patterns and groundwater quality with respect to groundwater Arsenic and Fluoride concentration in Lahore city and the associated health issues. The main goals of this study are;

- Relating human-induced land use and land cover changes to groundwater contamination in terms of increased Arsenic and Fluoride contamination.
- Estimation of the Associated Health Risk among the residents of the study area.

## 2. MATERIALS AND METHOD

### 2.1. Study Area

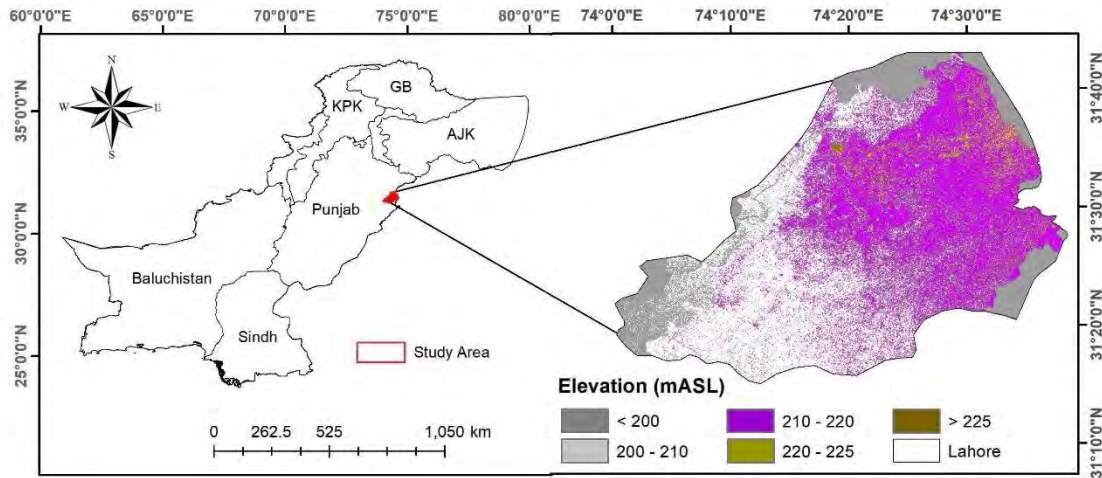
Lahore is one of the major cities of Pakistan and is ranked as the second largest city. It is located in Punjab province and lies between 24-37 °N and 62-75 °E, in the south-eastern region of Pakistan. Lahore falls within the alluvial plains of the south-flowing river Indus and its tributaries and is located on the eastern edge of the Ravi River. The city covers an area of approximately 1772 km and has a population of about 13,095,166 (Mushtaq et al., 2018). Lahore city experiences all four seasons of the year and is distinguished by cold winters and slurry-type summers and has an arid to semi-arid climate.

#### 2.2.1. Climate and geology of the study area

The province of Punjab is characterized by a subtropical continental and semiarid climate with hot and humid summers and cold winters. There exists variability in temperature range and rainfall depending on season-to-season changes. The average range of temperature lies between 120°C in winters and 340°C in summers and the average rainfall of about 575mm per year which can alter from 300 to 1200 mm per year (Abbas et al., 2022).

According to geological reports, Lahore consists of two aquifers, one shallow and the other deep aquifer (Bibi et al., 2015). These aquifers are composed of unconsolidated alluvial sediments approximately 400m thick, along with some amount of clay, sand, and silt. These underlying aquifers are separated by an aquitard.

Lahore is a densely populated area and with every passing year, the population is increasing. The population of the city is over 5.1 million with a 3.7% population growth rate (Abbas et al., 2022). This results in a number of issues among which the issue of the availability of clean drinking water is most important. The major source of drinking water in Lahore city is groundwater and the majority of the people rely on this source. Arsenic and Fluoride contamination in Lahore has been shown by different researchers and they have identified the relevant causes behind the contamination in their studies (Farooqi et al., 2007; Kumar et al., 2020b; Rehman et al., 2022). Among the different causes behind Arsenic and Fluoride contamination in the drinking water of Lahore, one major contributing factor is the human-induced intervention in the land use and land change pattern in the city.



**Figure 6:** Study Area, Lahore.

## 2.2. Data sets and method

### 2.2.1. Data acquisition

This research utilized Landsat data for the years 1995, 2006, 2011, 2016, and 2021, specifically Landsat 5-TM, Landsat 7-ETM+, and Landsat 8-OLI. The data was acquired from the official website of the United States Geological Survey (USGS). Remote sensing, a scientific technique for studying objects or phenomena without physical contact, was employed in this study. Remote sensing data, which includes images and metadata files, holds significant importance in analyzing land use and land cover changes at a large scale. To investigate such changes, satellite data was procured by downloading the data for a specific tile, namely tile 149 (Path)/38 (Row), as it corresponds to the geographic location of the study area. To ensure image clarity, only data with a cloud cover of less than 20% were selected for download.

**Table 3:** Remote sensing data

Year	Platform	Sensor	Date	Path/Row	Resolution	Cloud cover
1995	Landsat-5	TM	1995.5.06	149/38	30	<20%
2006	Landsat-7	ETM+	2006.5.23	149/38	30	<20%
2011	Landsat-7	ETM+	2011.5.21	149/38	30	<20%
2016	Landsat-8	OLI	2016.5.26	149/38	30	<20%
2021	Landsat-8	OLI	2021.5.24	149/38	30	<20%

### 2.2.2 Methodology

The following three main steps were used;

1. Land use land cover (LULC) classification
2. Accuracy assessment
3. Land use land cover (LULC) relationship and groundwater Arsenic and Fluoride relationship.
4. Human health risk assessment.

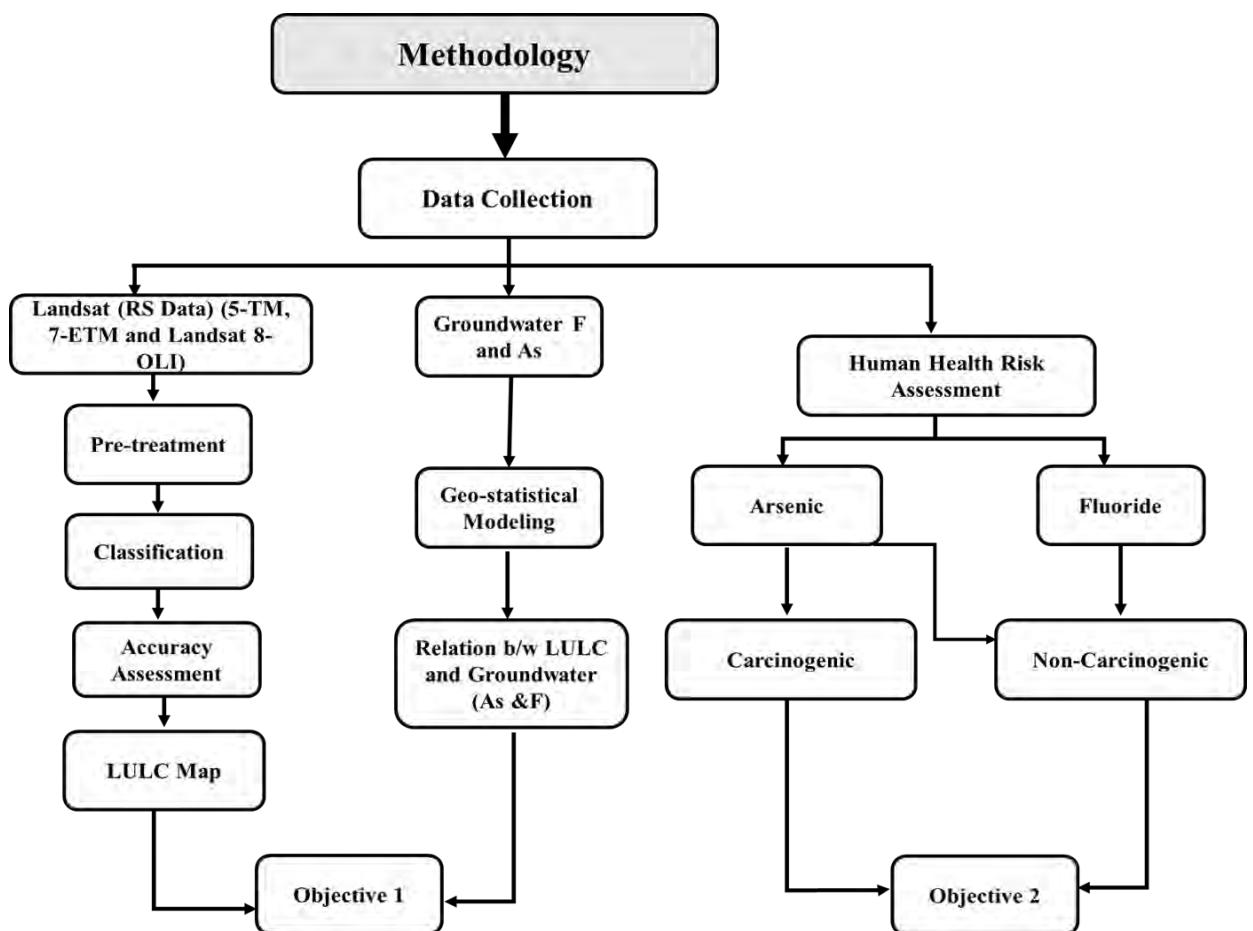


Figure 7: Methodology flow chart.

### 2.2.3 Data processing

The data obtained for the study was in its original, unprocessed form. In order to analyze changes in land use and land cover, the data needed to undergo some initial processing steps. These steps included correcting for atmospheric effects, adjusting the data for accurate measurements, and addressing specific issues with the Landsat 7-ETM+ sensor



that caused lines to appear in the images since 2003. By performing these pre-treatment processes, the data was prepared for further analysis and interpretation.

#### **2.2.4 Atmospheric and Radiometric correction**

To obtain accurate information from satellite images, it is necessary to rectify the effects of the Earth's atmosphere. This process is called atmospheric correction, and it enhances the images by removing atmospheric interference. In this study, atmospheric correction was performed on the acquired satellite images using QGIS software, allowing the images to be more effectively utilized for analysis.

Another important step is radiometric correction, which is used to reduce errors in satellite images caused by random factors. This correction process adjusts the digital numbers in the images to improve their accuracy. To carry out radiometric correction, specific information about the satellite sensor is needed, and this information is typically stored in the metadata file accompanying each image. In this study, the acquired Landsat images were subjected to radiometric correction using the SCP tool within the QGIS software.

#### **2.3. Land use land cover (LULC) classification and accuracy assessment**

In the classification process, we need to create training sites or regions of interest. These training sites are polygons that we draw to represent different classes or categories. They serve as samples for the software to learn and classify the image into those specific classes. There are different algorithms used for classification, such as the Maximum Likelihood algorithm and Support Vector Machines (SVM). The Maximum Likelihood algorithm classifies the image based on probabilities, considering the likelihood of each pixel belonging to a particular class. On the other hand, SVM is a non-parametric classifier that uses a learning algorithm to classify the image into different classes.

In order to analyze the changes in land use and land cover (LULC) during the study, the Landsat images were classified into different categories such as vegetation, fallow land, build-up areas, barren land, and water bodies. There are two main types of LULC classification methods: supervised and unsupervised. Unsupervised classification is a method where the software automatically classifies the image without requiring user input. On the other hand, supervised classification involves the user providing training samples from each class. These training samples are used to create a signature file. With

the training samples and signature file, the computer performs the classification for the entire image.

In this study, supervised classification was conducted using the Support Vector Machine algorithm with the help of ENVI software. This approach allowed for more accurate classification of the image into the desired land use and land cover categories.

After performing the classification, the next step was to assess the accuracy of the results. It is not possible to have ground truth information for every single pixel in the classified image. Therefore, random points were created within the classified image to serve as reference points. These reference points were divided into two sets of classes.

The first set of classes represented the land cover types as depicted in the classified image. The second set of classes acted as reference values and were based on the ground truth data provided by the researcher. To determine the accuracy, 40 reference pixels were generated for each classified image in this study. These reference points allowed for a comparison between the classified image and the ground truth data, providing a measure of how accurately the classification represented the actual land cover types.

#### **2.4. Groundwater Fluoride and Arsenic concentration data**

The data on the groundwater Fluoride and Arsenic concentration was obtained from already existing data. For the year 2006, the data of Lahore groundwater Fluoride and Arsenic concentration was taken in which a sum of 24 samples of groundwater were obtained from Kalalnwala area. Among these samples, 17 were obtained from shallow hand-pumped wells with depths ranging from 24 to 27 meters. Additionally, three samples were collected from electrically pumped tube wells specifically utilized for drinking water supply at depths between 60 and 90 meters. Furthermore, four samples were taken from electrically pumped irrigation wells with depths varying from 165 to 183 meters.

The data for the year 2011 was obtained from the study done in Lahore, in which a comparative study was done in three villages i.e., Kalalanwala (KLW), Manga Mandi (MM), and Shamki Bhattian (SKB). A total of 30 groundwater samples were gathered, with 20 obtained from shallow hand pumps installed at depths ranging from 24 to 36 meters. Additionally, 9 samples were collected from depths of 40 to 80 meters, while 1 sample was taken from a deep tube well with a depth of 80 to 200 meters.

For the year 2016, data for Arsenic concentration in the groundwater of Lahore was taken through blanket survey Four locations (Samada, Sarai Chimba, Kot Maiga, and Chah Fatehwala) were chosen along the Ravi River. Samples of groundwater were obtained from wells, including both hand pumps and boreholes, situated at various depths ranging from 24 to 80 meters. These wells were located approximately 2 kilometers away from the Ravi River.

Similarly, for Fluoride concentration, four sites in the districts of Lahore (Samada) and Kasur (Sari Chimba, Kot Maiga, and Chah Fatehwala) were taken. A total of 66 groundwater samples were gathered from existing wells, including both hand pumps and boreholes. The depths of these wells ranged from 24 to 76 meters, all of which were located within the unconfined aquifer.

For the year 2021, data on Fluoride and Arsenic concentration was obtained. A comprehensive initiative was undertaken to examine a total of 28,648 wells throughout the Punjab plains of Pakistan and India. The primary approach utilized field kits to assess the magnitude of the issue and gain insights into the underlying mechanisms. As part of this effort, groundwater samples were obtained from a subset of 712 wells specifically for laboratory analysis of Fluoride (F).

### **2.5 Relation between groundwater Fluoride, Arsenic and land use land cover change**

To determine the association between Fluoride and Arsenic concentration in groundwater and land use and land cover changes, geo-statistical methods were used. Geo-statistics is a methodology employed to analyze and predict values associated with spatiotemporal processes. It allows for the examination and estimation of values that are interconnected with the spatial and temporal dimensions. Geo-statistics aids in understanding the patterns, variability, and relationships within the data, thereby facilitating the analysis and prediction of values linked to spatiotemporal or spatial processes.

Inverse Distance Weighting (IDW) is a commonly used interpolation method in ArcGIS, a geographic information system (GIS) software. IDW in ArcGIS is used for spatially interpolating values from a set of known data points to estimate values at unknown locations within a study area. It assigns weights to nearby data points based on their distances from the prediction location, with closer points receiving higher weights. It is employed to create continuous maps or surfaces by interpolating point data. It can be used

to generate maps of various environmental and geospatial variables, enabling visual representation and analysis. IDW is a widely utilized algorithm for spatially interpolating point data, allowing for the estimation of values at locations where measurements were not taken. The method assumes that each measurement point has a local influence that diminishes with distance, and the strongest influences are in close proximity to the observed point.

In the study, using the inverse distance weighted (IDW) interpolation technique in ArcGIS 10.5.42, spatiotemporal surface maps were generated. IDW was chosen as the preferred interpolation technique because it ensured that the predicted values at unsampled locations fell within the range of the minimum and maximum values observed in the data.

The following equation was utilized for the application of IDW;

$$\text{Equation (1). } y(p) = \frac{\sum_{i=1}^n y_i / \beta k^c}{\sum_{i=1}^n 1 / \beta k^c}$$

Where;

- $y(p)$  = Interpolated value at the target location  $p$
- $y_i$  = Value of  $i$ th sample point
- $\beta k$  = Distance between the target location and  $i$ th point
- $n$  = Total number of samples
- $C$  = Power parameter that determines the weight assigned to each sample point

Following the aforementioned process, zonal statistical methods were used to assess the influence of land use/land cover on water quality parameters. Zonal statistical analysis is a statistical tool that computes the value for each zone based on data obtained from another dataset. In this particular study, the centroid of the union councils was transformed into point features. These point features facilitated the extraction of water quality values from interpolated raster, along with land-use/land cover data in tabular form. The "zonal statistics as table" tool in ArcGIS 10.5 was utilized to accomplish this task effectively.

### 2.5.2 Classification of groundwater Fluoride and Arsenic concentration

In order to calculate the concentration of groundwater Fluoride and Arsenic in different land use and land cover patterns that have been described earlier, the concentration data for the selected years were divided into five ranges for both Fluoride as well as for Arsenic. The ranges were kept in a way that all the values were being incorporated and thus an estimation with respect to the WHO standard can be made based on this classification. The ranges are as follows.

1. **Fluoride:** (i) <1mg/l, (ii) 1 to <1.5mg/l, (iii) 1.5mg/l to <3mg/l, (iv) <3mg/l to 5mg/l and (v) >5mg/l.
2. **Arsenic:** (i) <20 µg/l, (ii) 20 µg/l to <50 µg/l, (iii) 50 µg/l to <100 µg/l, (iv) 100 µg/l to 150 µg/l and (v) >150 µg/l.

## 2.6. Human health risk assessment

### 2.6.1 Exposure assessment

Human health risk assessment was done for the years 2006, 2011, 2016, and 2021. For Arsenic, both carcinogenic as well as non-carcinogenic risks were calculated. While for Fluoride, only non-carcinogenic risk was calculated since fluoride does not has carcinogenic effects. The health risk assessment was done on children below 6 years old for Fluoride and for Arsenic the age limit was set to greater than 16 years old. The oral mode was taken in this study to estimate the probability of individuals being exposed to high Arsenic and Fluoride in drinking water. For Arsenic, the chronic daily intake CDI through drinking water via oral pathway was determined using the following equation:

$$\text{Equation (2). } ADD_{ing} = \frac{C \times IR \times EF \times ED}{W \times T}$$

In the above equation,

- ADD is the average daily dose.
- C represents Arsenic concentration in water (µg/L)
- IR represents a water ingestion rate of 0.78 L/day for F and 2.5 L/day for As. (Ali et al., 2019; Shahab et al., 2018)
- ED is the exposure duration assumed to be 6 years for F and 16 years for As.
- EF shows the exposure frequency which is 365 days per year.
- BW is the body weight , which is 15kg for F and 72kg for As (Rasool et al., 2016).

AT is the average lifetime equal to 2190 days for F and 24575.45 days for As.

### **Non-carcinogenic risk**

The hazard quotient for non-carcinogenic risk was calculated using the following equation.

$$\text{Equation (3). } HQ = \sum \frac{DI}{RfD}$$

- HQ=Hazard Quotient
- DI=Daily Intake
- RfD=Reference Dose

The RfD for Arsenic is 0.0003mg/kg/day. While RfD for Fluoride is 0.06mg/kg/day as calculated by USEPA (2011).

### **Carcinogenic risk**

Carcinogenic risk for Arsenic was calculated through the following equation.

$$\text{Equation (4) } CR = \sum DI \times SF$$

- DI=Daily Intake
- SF= Cancer Slope Factor equal to 1.5mg/kg/day (USEPA 2011)

### 3. RESULTS AND DISCUSSION

#### 3.1 Accuracy assessment of land use and land cover (LULC)

The accuracy Assessment for the LULC maps was determined and the results are shown below in the table.

**Table 4:** Accuracy assessment of the classified maps for the year 1995, 2006, 2011, 2016 and 2021.

<b>Years</b>	<b>User Accuracy (%)</b>	<b>Producer Accuracy (%)</b>	<b>Overall Accuracy (%)</b>	<b>Kappa coefficient</b>
<b>1995</b>	94.65	91.96	93.03	0.9
<b>2006</b>	94.78	94.9	93.28	0.91
<b>2011</b>	95.3	93.39	94.03	0.91
<b>2016</b>	96.54	94.4	93.8	0.9
<b>2021</b>	95.5	92.32	95.91	0.91

It can be seen that the overall accuracy is above 90%, which is an indication that the accuracy is high and the maps are accurate.

#### 3.2 Land use and land cover changes

##### 3.2.1. Land use land cover change 1995

The findings pertaining to Land Use Land Cover (LULC) in 1995 suggest that a considerable part of the land in the study area was dedicated to agricultural activities, including both crop cultivation and fallow land. Specifically, agriculture accounted for the majority of land coverage. Furthermore, approximately 78% of the area was characterized by vegetation, making it the dominant land cover category for that year. In contrast, barren land encompassed around 11% of the study area, representing a significant proportion as well. Aside from agricultural and barren lands, approximately 10% of the area was occupied by built-up structures, signifying human settlements and urban development. Additionally, water bodies accounted for nearly 1% of the total land area in Lahore and this is majorly the portion of river Ravi.

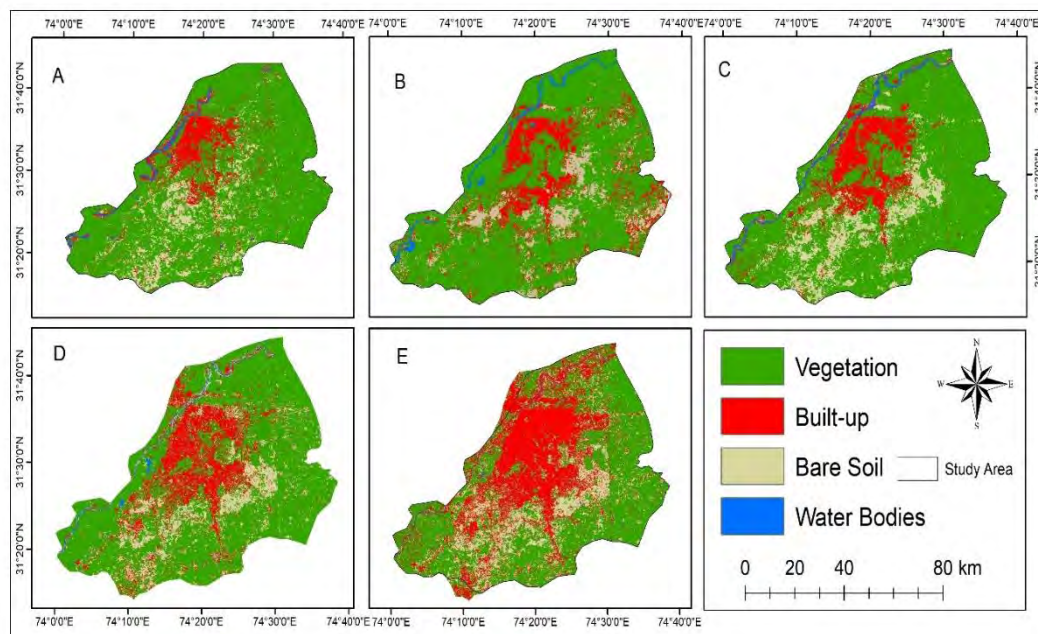
##### 3.2.2. Land use land cover change 2006

Based on the findings, the year 2006 revealed that a significant portion of Lahore city was characterized by vegetation, constituting approximately 72% of the total area. This

indicates the presence of greenery and natural cover throughout the city during that period. In contrast, around 14% of the study area was identified as barren land, which suggests areas with limited vegetation or barren terrain unsuitable for significant vegetation growth. The built-up areas accounted for approximately 12% of the entire city area in 2006. This category represents urbanized and developed regions, including residential, commercial, and industrial areas. While water bodies, such as rivers, lakes, and ponds, occupied about 2% of Lahore in 2006.

### 3.2.3 Land Use Land Cover Change in 2011

The findings pertaining to land use land cover change in Lahore city for the year 2011 unveiled that within the overall spatial extent of the city, vegetation cover accounted for approximately 64% of the total land area, signifying the dominant presence of thriving plant communities. Conversely, approximately 19% of the total city area exhibited characteristics of barren land, denoting regions with limited or absent vegetation cover. The built-up areas, encompassing around 16% of the city's territory, epitomized the spatial expansion of urbanization and human settlements. These developed zones encompassed residential, commercial, and industrial infrastructure, exemplifying the transformation of natural landscapes into urbanized environments. The water component, accounting for 2% of the city's land area, predominantly corresponded to the presence of the river Ravi, which traverses the city.



**Figure 8:** LULC maps, (A)1995, (B) 2006, (C) 2011, (D) 2016, E 2021

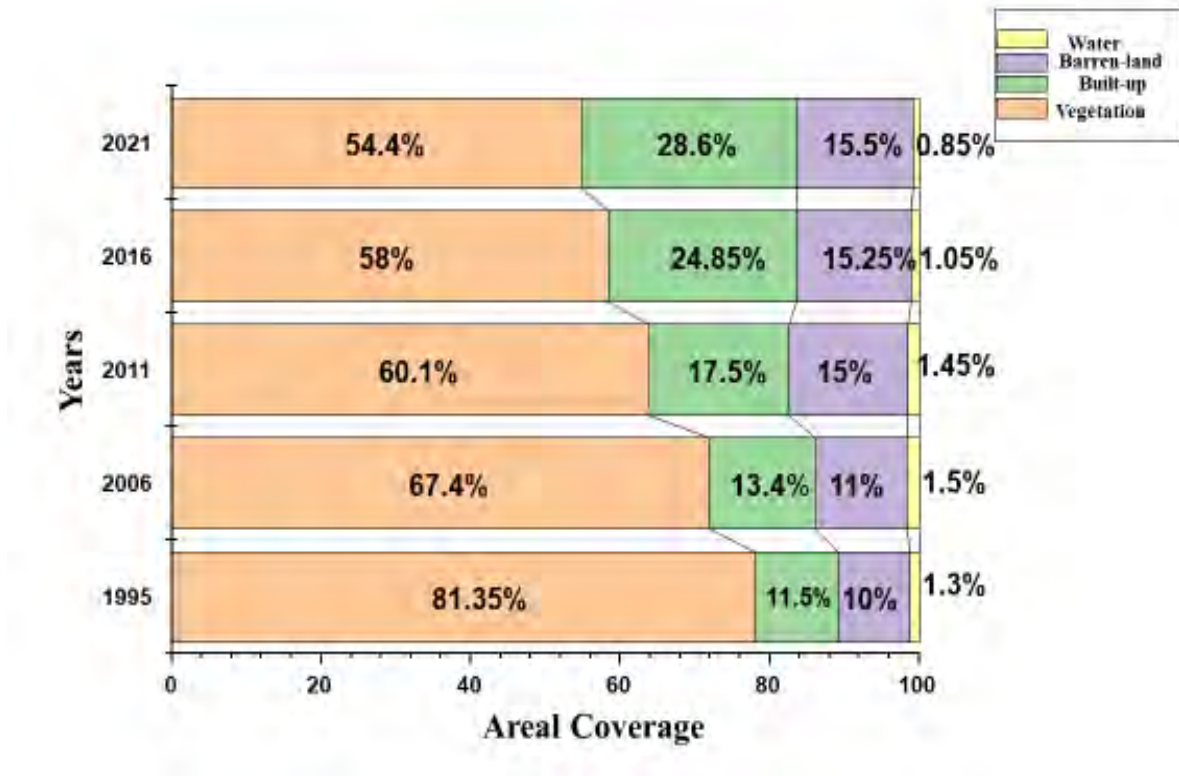


#### **3.2.4. Land Use Land Cover Change in 2016**

The land use land cover analysis conducted in 2016 revealed distinct patterns regarding the distribution of different land cover classes in the study area. The predominant land cover class was vegetation, occupying the largest portion of the land, with a significant percentage value of 58. Following vegetation, the next prominent land cover class was barren land, encompassing approximately 25% of the total study area. This category represents areas characterized by limited or absent vegetation, such as deserts, rocky terrains, or land affected by degradation. The built-up areas, consisting of residential, commercial, and industrial infrastructure, constituted around 15% of the total area. This indicates the extent of urban development and human settlements within the studied region during the year 2016. Water bodies, including rivers, lakes, and ponds, accounted for approximately 1% of the total area in 2016.

#### **3.2.5 Land Use Land Cover Change in 2021**

The land use land cover analysis conducted in 2021 unveiled distinct patterns in the distribution of land cover classes within the study area. Vegetation emerged as the dominant land cover class, encompassing the largest land portion, representing a substantial percentage of 55. Following vegetation, barren land emerged as the next significant land cover class, occupying approximately 29% of the total study area. Barren land comprises regions characterized by limited or absent vegetation, such as deserts, rocky terrains, or areas affected by degradation. The built-up areas, which include residential, commercial, and industrial infrastructure, constituted around 16% of the overall land area. Water bodies, such as rivers, lakes, and ponds, accounted for a modest proportion of approximately 1% of the total area in 2021.



**Figure 9:** Areal coverage of different LULC in different years.

From the results above it can be seen that the area coverage for vegetation is decreasing from 1995 to 2021 while the built-up areas in the city are increasing. Lahore has experienced rapid urbanization and population growth over the past few decades. As the city expands, more land is converted for residential, commercial, and industrial purposes. This leads to the destruction of vegetation and the conversion of green spaces into concrete setups (Brindha & Schneider, 2019). Increased urbanization often results in deforestation and tree loss. Trees are cut down to make way for infrastructure development, including roads, buildings, and housing projects.

The expansion of road networks, construction of buildings, and the development of urban infrastructure contribute to the reduction of vegetation coverage. These activities require the clearing of land, including forests, parks, and green areas, resulting in a decline in vegetation cover. An observable trend from 1995 to 2021 reveals a noteworthy increase in barren land within the region. This phenomenon can be attributed to an ongoing process characterized by the abandonment of agricultural activities on arable lands, subsequently resulting in their conversion into built-up areas. This process involves the abandonment of farming activities on agricultural lands, leading to a net gain in barren

areas. Subsequently, these abandoned lands are converted into built-up areas, further contributing to the expansion of barren land (Imran & Mehmood, 2020). The areal coverage of water in the city shows that it is nearly constant.

### 3.3 Change in land use and land cover

According to the results shown in Table 5, land use land cover showed variation in different land cover classes. Initially, in 1995, vegetation covered 1627sq. km of the total area of the city of Lahore. In the first eleven years of duration, the vegetation decreased by 279 km, while during the second time period, it was further decreased by 146 km. From 2011 to 2016, the 42km area of vegetation was reduced and from 2016 to 2021 72km of vegetation area was decreased. The net change that occurred in vegetation during the study period was noted as -26.95% in Lahore. Similarly, built-up was increased during the first eleven years (1995-2006) by 38 km from 230km and it was increased by 82 km during the year 2006 to 2011. In the next 5 years (2011 to 2016), an increase of 147km was observed in the built-up area. Likewise, from 2016 to 2021, the area coverage of built-up further increased. So the net change in built-up was noted as +17.1%.

The results for barren land showed that in the first time period of 1995 to 2006, the portion of barren land increased up to 30km from 200km, followed by another increasing trend in the year 2006 to 2011 by 70km. Then from 2011 to 2016 the change in the barren land was observed to be 5km more than the previous year. A further increase of 5km was shown in the year 2016 to 2021. The net change in barren land was found to be + 5.5%. In case of water, the variation throughout the study time was observed to be less fluctuating with a net change of -0.45%.

**Table 4:** Net change (in %age) in LULC from the year 2006 to 2021.

<b>Class</b>	<b>1995 (%)</b>	<b>2006 (%)</b>	<b>2011 (%)</b>	<b>2016 (%)</b>	<b>2021 (%)</b>	<b>Net Change (%)</b>
<b>Vegetation</b>	81.35	67.4	60.1	58	54.4	-26.95
<b>Built-up</b>	11.5	13.4	17.5	24.85	28.6	+17.1
<b>Barren-Land</b>	10	11.5	15	15.25	15.5	+5.5
<b>Water</b>	1.3	1.55	1.45	1.05	0.85	-0.45

### 3.4 Groundwater Fluoride and Arsenic concentration

The concentration of Fluoride and Arsenic in the groundwater of Lahore in the years 2006, 2011, 2016, and 2021 showed a noticeable variation in different land use and land cover class. In every year the concentrations are seen to be increasing from the WHO standard value. The table below shows the variation in the concentration of Fluoride in the groundwater of Lahore. The concentration of Fluoride along with mean values in the different land use and land cover classes is depicted in the following table.

**Table 5:** Fluoride (mg/l) data for groundwater in different years, in the respective LULC classes.

<b>Year</b>	<b>Class</b>	<b>Min</b>	<b>Max</b>	<b>Mean</b>	<b>SD</b>
<b>2006</b>					
	Vegetation	0.57	14.52	4.44	±2.18
	Built-up	0.22	21.53	4.43	±1.62
	Barren Land	0.66	12.21	5.07	±1.40
	Water	0.28	20.53	4.79	±2.13
<b>2011</b>					
	Vegetation	0.20	1.82	0.95	±0.44
	Built-up	0.20	5.15	0.85	±0.36
	Barren Land	0.14	5.15	0.72	±0.30
	Water	0.24	5.06	0.89	±0.33
<b>2016</b>					
	Vegetation	1.0026	16.88	4.15	±2.08
	Built-up	1.0031	17.71	3.50	±1.57
	Barren Land	1.0032	16.90	3.64	±1.85
	Water	1.2158	10.36	3.70	±1.87
<b>2021</b>					
	Vegetation	2.35	11.32	5.56	±0.97
	Built-up	1.39	13.64	5.46	±0.83
	Barren Land	1.17	17.21	5.32	±1.48
	Water	1.02	14.86	5.66	±1.17

Similarly, the concentration of Arsenic in the groundwater of Lahore in different LULC classes is shown in the following table.

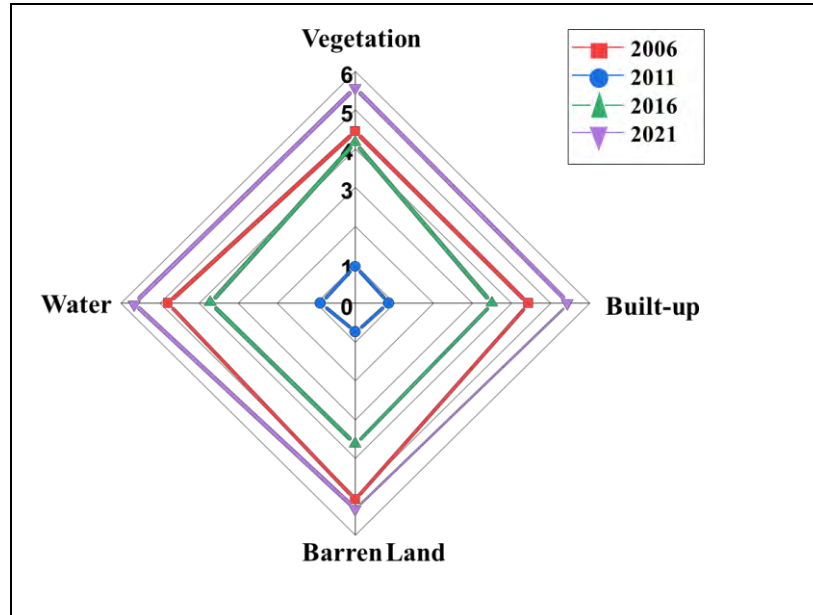
**Table 6:** Arsenic ( $\mu\text{g/L}$ ) data of groundwater in different years, in the respective LULC classes.

<b>Year</b>	<b>Class</b>	<b>Min</b>	<b>Max</b>	<b>Mean</b>	<b>SD</b>
<b>2006</b>					
	Vegetation	7.68	170.53	102.38	$\pm 24.34$
	Built-up	4.48	671.06	111.84	$\pm 53.69$
	Barren Land	6.85	590.19	106.66	$\pm 83.55$
	Water	3.55	641.92	54.27	$\pm 59.47$
<b>2011</b>					
	Vegetation	31.96	105.19	64.10	$\pm 16.25$
	Built-up	5.77	498.32	69.29	$\pm 24.70$
	Barren Land	10.12	452.72	63.75	$\pm 24.43$
	Water	12.12	146.73	58.53	$\pm 18.49$
<b>2016</b>					
	Vegetation	0.02	507.98	58.05	$\pm 44.49$
	Built-up	0.25	544.69	53.08	$\pm 45.33$
	Barren Land	0.14	459.38	57.70	$\pm 54.09$
	Water	1.61	108.73	30.04	$\pm 22.84$
<b>2021</b>					
	Vegetation	18.11	463.25	123.57	$\pm 40.81$
	Built-up	17.32	544.46	145.22	$\pm 71.39$
	Barren Land	18.28	484.88	141.71	$\pm 52.14$
	Water	25.26	355.49	127.49	$\pm 54.35$

### 3.5 Groundwater Fluoride and Arsenic concentration in different classes

#### 3.5.1 Fluoride

The results obtained from the study on the concentration of fluoride in the groundwater of Lahore, specifically concerning the selected land use classes, reveal a consistent and concerning net-increasing trend from 2006 to 2021. This finding highlights the progressive contamination of groundwater by fluoride over the studied period. Notably, the highest concentration of fluoride was found in water, indicating a significant risk for communities reliant on groundwater as a primary drinking water source. The presence of elevated fluoride levels in water bodies suggests potential contamination from various sources, such as industrial effluents, agricultural runoff, and domestic wastewater. This high fluoride content is directly responsible for tooth discoloration and bone deformation observed among residents.



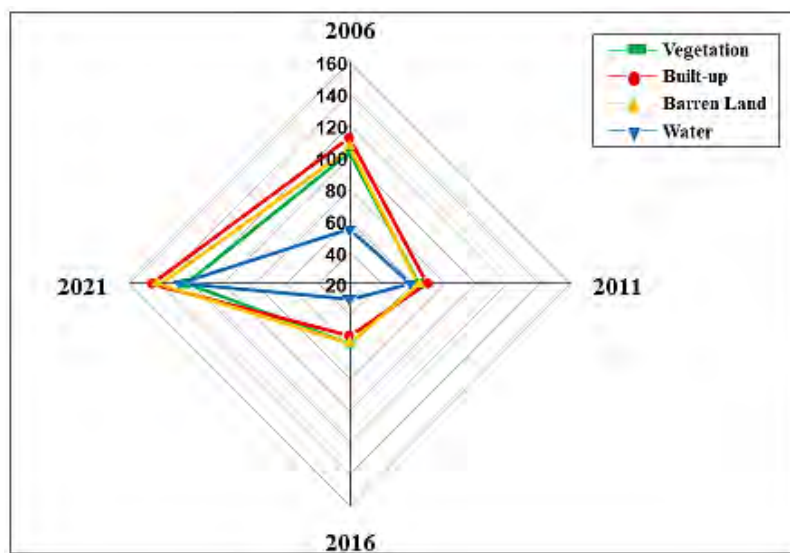
**Figure 10:** Fluoride mean in different LULC.

Moreover, the study observed elevated fluoride concentrations in vegetation, indicating the potential for fluoride accumulation through root uptake from groundwater or atmospheric deposition. This finding raises concerns regarding the consumption of crops

and other edible plants grown in areas with high fluoride concentrations, as they can serve as pathways for human exposure. Additionally, the study identified comparatively lower fluoride concentrations in built-up and barren land areas. However, the presence of fluoride in these areas indicates that anthropogenic activities, such as the use of fluoride-containing construction materials and industrial discharges, may contribute to localized contamination. Strict regulations, effective waste management practices, and the adoption of eco-friendly construction techniques are crucial for minimizing fluoride contamination in these areas and ensuring the well-being of urban populations.

### 3.5.2 Arsenic

The systematic analysis of arsenic concentration in the groundwater of Lahore reveals a net increasing trend from 2006 to 2021. Built-up areas displayed the highest concentrations, succeeded by barren land, water, and vegetation. The rising trend of arsenic concentration over the studied period raises significant concerns about the groundwater quality in Lahore. This increasing contamination may be attributed to various factors, including industrial activities, agricultural practices, and natural geological sources.



**Figure 11:** Arsenic mean in different LULC.

Built-up areas exhibited the highest concentrations of arsenic, indicating the influence of anthropogenic activities. Industrial discharges, improper waste disposal, and the use of arsenic-containing materials in construction may contribute to the contamination in these

areas. Barren land also showed elevated concentrations of arsenic, suggesting the contribution of natural geological sources. Water sources exhibited lower arsenic concentrations compared to built-up areas and barren land. However, the presence of arsenic in water raises concerns about its potential impact on human health, particularly through drinking water consumption. Vegetation showed the lowest concentrations of arsenic among the selected classes.

### **3.6 Relation between groundwater Fluoride, Arsenic concentration, and land use land and land cover change**

The study aimed to investigate the interrelationships among groundwater Fluoride, Arsenic concentration, and land use land cover change through the utilization of the Inverse Distance Weighted (IDW) interpolation technique. By adhering to the predetermined concentration ranges for Fluoride and Arsenic, as outlined in the methodology, the application of IDW effectively highlighted regions exhibiting high concentrations of both elements within the groundwater. Moreover, the IDW maps accurately represented all values falling within the defined concentration range for arsenic and fluoride, offering comprehensive visual insights into their spatial distribution patterns in the different classes of the study area. After this, the zonal statistical table was used to find the relationship between land use and land cover changes, and groundwater Fluoride and Arsenic concentration.

#### **3.6.1. Fluoride**

According to the results obtained from the zonal statistics analysis, it has been determined that the concentration of fluoride exhibits distinct patterns across different land classes over a span of several years. In the year 2006, the barren land displayed the highest fluoride concentration, reaching a value of 5.0709 mg/L. Following closely behind were the water class with a value of 4.79 mg/L, the vegetation class with 4.44 mg/L, and lastly the built-up areas with a relatively lower value of 4.43 mg/L, indicating a comparatively lesser fluoride presence in this particular land category.

Shifting the focus to the subsequent year, 2011, a different trend emerged in the fluoride distribution. The vegetation class exhibited the highest concentration of fluoride with a value of 0.95 mg/L, surpassing other land classes. Water ranked second with a value of 0.89 mg/L, while the built-up areas showed a fluoride concentration of 0.85 mg/L. The



barren land class displayed the lowest concentration among these categories, registering a value of 0.72 mg/L.

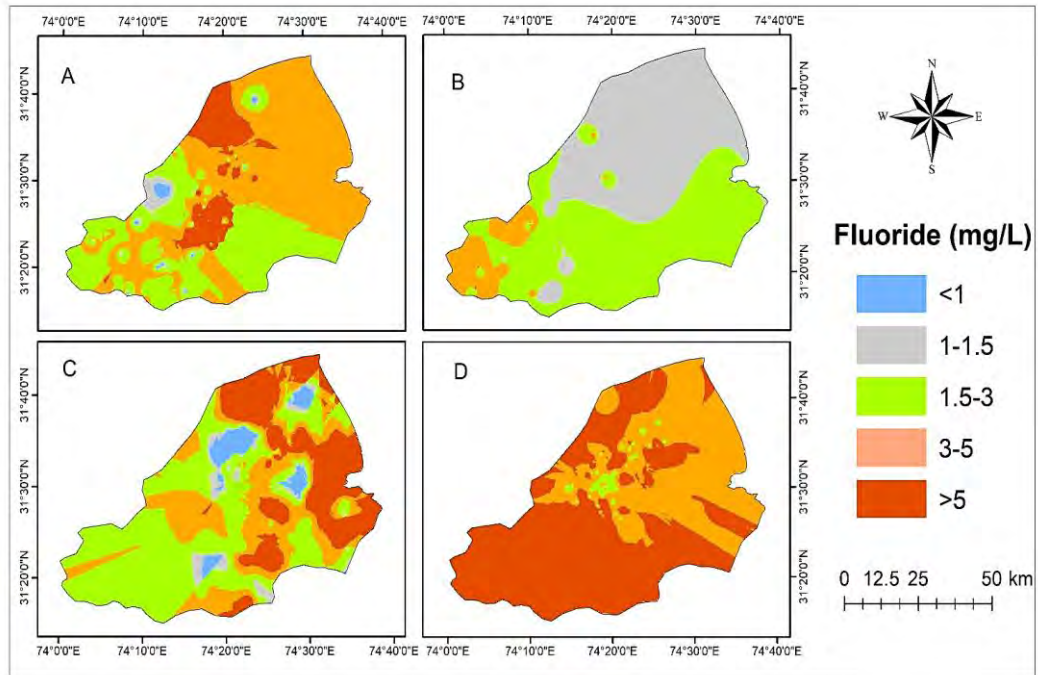
**Table 7:** Zonal statistical table for Fluoride (mg/L).

<b>Class</b>	<b>2006</b>	<b>2011</b>	<b>2016</b>	<b>2021</b>
<b>Vegetation</b>	4.44	0.95	4.15	5.56
<b>Build-up</b>	4.43	0.85	3.50	5.46
<b>Barren land</b>	5.07	0.72	3.64	5.32
<b>Water</b>	4.79	0.89	3.70	5.66

Moving forward to the year 2016, it became apparent that the vegetation class retained its position as the land category with the highest fluoride concentration, with a value of 4.15 mg/L. The water class followed closely behind, exhibiting a fluoride value of 3.70 mg/L. Meanwhile, the barren land and built-up areas displayed fluoride concentrations of 3.6 mg/L and 3.9 mg/L, respectively, showing relatively similar levels. Finally, focusing on the most recent year analyzed, 2021, the water class demonstrated the highest fluoride concentration with a value of 5.66 mg/L. The vegetation class ranked second with a fluoride value of 5.56 mg/L, followed by the built-up areas with a value of 5.46 mg/L. The barren land class exhibited a fluoride concentration of 5.32 mg/L, placing it last among the analyzed land categories in terms of fluoride presence.

In summary, the zonal statistics analysis reveals varying fluoride concentrations across different land classes over the course of several years. The barren land class displayed the highest concentration in 2006, while the vegetation class showcased the highest fluoride values in 2011, 2016, and 2021. Water consistently exhibited high fluoride concentrations across multiple years, with the built-up areas consistently displaying relatively lower fluoride values in comparison.

Following is the graphical representation of the above results.



**Figure 12:** Fluoride geo-statistical results, (A) 2006, (B) 2011, (C) 2016, (D) 2021.

The observed elevation of fluoride concentration in vegetation over the course of years can be attributed to the widespread utilization of various fertilizers and pesticides containing fluoride. These substances, when applied to the soil, can leach into the groundwater, leading to an accumulation of fluoride over time. One prominent contributor to this issue is the excessive application of phosphatic fertilizers, which are commonly employed to enhance crop yield. These fertilizers have emerged as a significant anthropogenic source of fluoride contamination in groundwater, as documented by Kundu and Mandal (2008). The application of these fertilizers releases fluoride ions into the soil, which can then percolate into the groundwater, thereby increasing its fluoride concentration.

Furthermore, the reliance on irrigation practices necessitates a constant supply of water, and the utilization of water with high fluoride content for irrigation purposes represents another potential cause for the elevated fluoride levels in the groundwater under the vegetation class. The use of high fluoride-containing water for irrigation purposes introduces additional fluoride into the soil, which can subsequently infiltrate the underlying groundwater, contributing to its elevated fluoride concentration consequently, this practice also contributes to the contamination of groundwater with fluoride, as highlighted by Pettenati et al. (2013). The excessive extraction of groundwater for

irrigation purposes can intensify the dissolution of fluoride from geological formations, such as rocks and minerals containing fluoride compounds (Kumar et al., 2016). This accelerated dissolution leads to an increased fluoride concentration in the groundwater, particularly in areas where the underlying geological composition is rich in fluoride-bearing minerals.

Water class consistently exhibited high fluoride concentrations across multiple years. The high concentration of fluoride in the groundwater under the water class can be attributed to different reasons. One major reason is the waste water discharge into the surface water. Improperly treated or untreated wastewater discharge can also introduce fluoride into surface water bodies. If this water subsequently infiltrates into the groundwater, it can lead to increased fluoride concentrations. Industries, domestic sewage, and agricultural runoff can be potential sources of fluoride-containing wastewater (Yasar et al., 2021). Furthermore, the natural weathering and erosion of rocks and minerals can contribute to the release of fluoride into the groundwater. Over time, water flowing over fluoride-containing geological formations can dissolve fluoride ions and transport them into the groundwater system, resulting in increased fluoride concentrations (Abbas et al., 2015). Groundwater recharge rate also plays an important role in determining the concentration of contaminants in the groundwater. Recharge refers to the process by which water from precipitation or surface water infiltrates into the groundwater system. The rate of recharge influences the dilution of contaminants, including fluoride, in groundwater. If the recharge rate is low, such as in arid or semi-arid regions, there may be limited freshwater input to dilute the fluoride, potentially leading to higher fluoride concentrations in the groundwater (Ling et al., 2022).

For the year 2006, barren land showed high fluoride concentration in the groundwater under it. This is due to the reason that in barren and arid regions, the limited availability of surface water and lower rainfall rates lead to reduced dilution of contaminants, including fluoride, in the groundwater. With less water percolating through the subsurface, there is limited flushing of fluoride from the aquifer, resulting in its accumulation over time. The absence of vegetation also reduces the uptake of water through transpiration, which could further contribute to the lower flushing of fluoride (Ashraf et al., 2022). Moreover, in barren land, high evaporation rates can result in the concentration of dissolved minerals, including fluoride, in the remaining water. As water

evaporates from the surface, the concentration of fluoride increases, which can then infiltrate into the underlying groundwater, elevating its fluoride concentration (Ismail & Ahmed, 2022).

The least value of fluoride in the built-up class is because of the reduced geological sources in this land class. Built-up areas typically have limited exposure to geological formations that naturally contain fluoride-rich minerals. As a result, there may be a lower prevalence of fluoride sources in the vicinity, reducing the likelihood of fluoride contamination in the groundwater (Rahman et al., 2018). Secondly, Built-up areas are characterized by extensive surface infrastructure, including roads, buildings, and pavement. These impermeable surfaces limit the direct interaction between rainfall and the underlying soil, reducing the infiltration of fluoride-contaminated water into the groundwater. The presence of impervious surfaces can act as a barrier, preventing the percolation of contaminants into the aquifer (Abbas et al., 2015).

### 3.7.2. Arsenic

According to the zonal statistical table for Arsenic concentration, the year 2006 revealed that the highest level of Arsenic was observed in the built-up class, reaching a value of 111.8394  $\mu\text{g/L}$ . Following closely was the barren land class, exhibiting a value of 106.6606  $\mu\text{g/L}$ . Comparatively lower concentrations were found in the vegetation class, measuring up to 102.37  $\mu\text{g/L}$ , while water displayed the least value of 54.26  $\mu\text{g/L}$  among the classes. Similarly, in 2011, the built-up class maintained its dominance with a value of 69.29  $\mu\text{g/L}$ , followed by vegetation at 64.09  $\mu\text{g/L}$ , barren land at 63.75  $\mu\text{g/L}$ , and water at 58.53  $\mu\text{g/L}$ , consecutively.

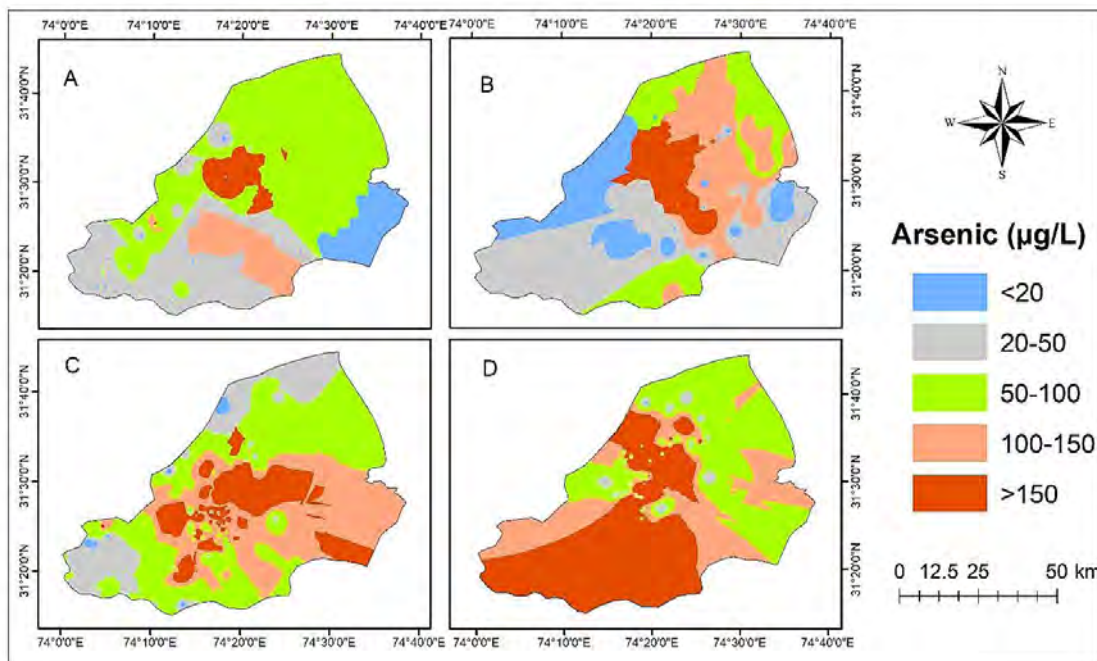
**Table 8:** Zonal statistical table for Arsenic ( $\mu\text{g/L}$ ).

<b>Class</b>	<b>2006</b>	<b>2011</b>	<b>2016</b>	<b>2021</b>
<b>Vegetation</b>	102.37	64.09	53.04	123.56
<b>Build-up</b>	111.83	69.29	58.07	145.22
<b>Barren land</b>	106.66	63.75	57.69	141.70
<b>Water</b>	54.26	58.53	30.04	127.48

In the year 2016, the zonal statistical analysis revealed elevated Arsenic concentrations within various land cover classes. Once again, the built-up class exhibited a high value of 58.07  $\mu\text{g/L}$ , indicating the highest Arsenic level. The barren land class followed closely with a value of 57.69  $\mu\text{g/L}$ , while the vegetation class displayed a concentration of 53.04  $\mu\text{g/L}$ . Lastly, the water class exhibited a lower value of 30.04 among the considered categories.

Similarly, the results from 2021 corroborated these findings, demonstrating persistent high Arsenic levels in specific land cover categories. The built-up category continued to exhibit a notably elevated value of 145.22  $\mu\text{g/L}$ , confirming it as the class with the highest Arsenic concentration. The barren land class followed closely with a value of 141.70  $\mu\text{g/L}$ , while water and vegetation displayed respective values of 127.48  $\mu\text{g/L}$  and 123.56  $\mu\text{g/L}$ , representing relatively lower Arsenic levels. This consistent trend across both years indicates the higher presence of Arsenic in built-up and barren land classes, while vegetation and water classes showcased comparatively lower levels.

Following is the graphical representation of the above results.



**Figure 13:** Arsenic geo-statistical results, (A) 2006, (B) 2011, (C) 2016 and (D) 2021.

The reasons behind the increased concentration of Arsenic in the built-up class can be majorly attributed to the fact that the built-up class has more ongoing anthropogenic

activities, which includes a number of industrial activities, residential and commercial activities, construction process, and many other developmental activities. These activities in one way or another releases different waste effluents which are either directly or indirectly entering the groundwater. These effluents include a number of harmful contaminants, which when enter the groundwater, they cause changes in the natural geological setup of the groundwater. Industrialization has significantly impacted Pakistan, and it is evident that groundwater in the region has been noticeably enriched with arsenic, reaching levels of up to 2.4 mg L<sup>-1</sup> (Farooqi et al., 2008).

Urban areas exhibit distinct characteristics with an abundance of impermeable surfaces resulting from the proliferation of buildings, roads, and associated infrastructure (Paul et al., 2018). The transformation of natural landscapes into urbanized environments significantly alters the hydrological dynamics, leading to diminished water infiltration and limited recharge capacity. As a consequence, the intricate groundwater circulation system undergoes substantial modifications, compromising its ability to effectively dilute and transport groundwater pollutants (Jia et al., 2018). Moreover, the escalating water demands driven by rapid urban population growth pose considerable challenges in ensuring an adequate and sustainable water supply for urban inhabitants. This issue becomes particularly pronounced in regions such as Europe and Asia, where the combination of intense urbanization and excessive groundwater pumping has contributed to the amplification of arsenic content within groundwater reserves (Brindha & Schneider, 2019; Minnig et al., 2018). Pakistan is among the rapidly urbanizing countries in South Asia and it is estimated that over the years the population living in the urban areas has increased from 17% in 1951 to 37% in 2010. and to 39.75 in 2017 (Jia et al., 2018). Lahore is one of the rapidly urbanizing cities of Pakistan with a population of 6.3 million in 1998, which increased to 111.2 million in 2017 having an annual growth of 3% (Imran & Mehmood, 2020). In recent decades, the city has undergone remarkable advancements, experiencing extensive development. Factors such as industrialization and the provision of enhanced residential amenities have played a pivotal role in stimulating the city's urban expansion and contributing to a steady rise in economic growth (Mumtaz et al. 2020). The expanding urban population exacerbates the degradation of groundwater quality, exacerbated by various factors including the infiltration of contaminants from landfill leachate and the application of fertilizers in green spaces (Hua et al., 2020).

Barren land is the second class where there is high concentration of Arsenic. This the class that actually is a transition from vegetation towards built-up. It is the intermediate class. For different reasons, including developmental causes, the vegetative land cover is converted to barren lands which are later on converted into built-up class. This land cover class has ongoing construction and commercial activities, industrial processes and other developmental works which leads to increasing concentration of different contaminants that alter the quality of underneath groundwater. Moreover, geological causes can also contribute to increasing Arsenic concentration in the groundwater. The hydrogeological properties of the barren land, such as low permeability or restricted groundwater flow, can lead to stagnant groundwater conditions. This stagnant water may undergo chemical reactions with the surrounding geological materials, resulting in the release of arsenic into the groundwater (Hua et al., 2020). Redox reactions occurring within the geological formations underlying the barren land can contribute to the release of arsenic into the groundwater. Changes in the oxygen levels and pH of groundwater can facilitate the dissolution and mobilization of arsenic from the geological materials.

The groundwater underlying vegetation and water exhibits relatively lower concentrations of arsenic compared to built-up and barren land. However, even these land classes display noticeable levels of arsenic concentration. The presence of arsenic in the vegetation class can be attributed to the utilization of agricultural inputs such as fertilizers, pesticides, and other products containing high concentrations of toxic contaminants. These substances can leach into the soil and subsequently affect the geochemistry of groundwater, leading to an increase in arsenic release and concentration. The leachable concentration of arsenic (As) in fertilizer samples collected from the area ranged from 7 to 10 mg/kg. (Farooqi et al., 2007).

Furthermore, the increasing agricultural demands in the region have resulted in the installation of additional tube wells for groundwater extraction. This excessive extraction leads to decreased groundwater dilution and increased saturation, thereby contributing to elevated arsenic concentrations in the groundwater (Li et al., 2021).

Over time, the landscape in Lahore has witnessed a shift from vegetation-dominated areas to built-up and barren land. As a result, the area coverage of vegetation has diminished, and built-up and barren land have emerged as significant sources of groundwater arsenic

contamination. Consequently, the built-up and barren land classes exhibit comparatively higher levels of arsenic concentration in the groundwater.

The river Ravi stands as the primary water body within the urban landscape of Lahore. The findings reveal that the water class exhibits the lowest concentration of groundwater arsenic. This observation suggests that the groundwater beneath the river Ravi has experienced limited modifications, with minimal disruptions to its geochemical composition.

### 3.8. Human health risk assessment

#### 3.8.1. Fluoride

The human health risk assessment done for Fluoride showed that the value of the hazard quotient has been increasing from the year 2006 to the year 2021.

**Table 9:** Human health risk assessment for Fluoride.

Years	ADD(mg kg <sup>-1</sup> day <sup>-1</sup> )			Non Carcinogenic Risk		
	Min	Max	Mean	Min	Max	Mean
2006	0.00624	0.494	0.16562	0.104	8.233333	3.012038
2011	0.0312	0.78	0.199808	0.52	13	3.330137
2016	0.02808	0.91	0.231676	0.468	15.16667	3.861263
2021	0.294667	14.82	2.512326	0.01768	14.82	4.512326

The hazard quotient (HQ) values, determined for children below the age of 6 years who were exposed to fluoride through the oral pathway of drinking water ingestion, exhibited a notable elevation surpassing the established threshold of 1, for the years 2006, 2011, 2016, and 2021. The World Health Organization (WHO) sets this threshold as an indication of increased health risks.

The table provided reveals a progressive trend in the HQ values, commencing with a value of 3.01 in 2006, which subsequently increased to 3.33 in 2011, further escalating to 3.86 in 2016, and reaching its highest recorded value of 4.5 in 2021. The consistent increase in hazard quotient (HQ) values can be attributed to the rising concentration of



fluoride in groundwater over time. Prolonged exposure to high fluoride levels in drinking water amplifies the potential health risks, especially for vulnerable populations. The heightened risk posed by excessive fluoride exposure in children can manifest in various health problems, including fluorosis, anorexia, salivation, tachycardia, stiffness, and restlessness, as reported by Sahu et al. (2016).

### 3.8.2. Arsenic

Human health risk assessment done for Arsenic in the groundwater of Lahore for the years 2006, 2011, 2016 and 2021 showed a consistent increase in the health hazards, carcinogenic as well as non-carcinogenic.

**Table 10:** Human health risk assessment for Arsenic.

Years	ADD (mg kg <sup>-1</sup> day <sup>-1</sup> )			Non-Carcinogenic Risk			Carcinogenic Risk		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
2007	0.000031	0.0833	0.0026	0.115	277.77	8.93	0.000052	0.125	0.0040
2011	0.00034	0.0227	0.0030	1.157	75.81	10.30	0.00054	0.034	0.0046
2016	0	0.0190	0.0033	0	63.42	11.28	0	0.028	0.0050
2021	0.000417	0.0201	0.0039	1.388	67.12	13.03	0.00062	0.030	0.0058

The Hazard quotient (HQ) values, which indicate the non-carcinogenic risk, exhibit an upward trend from 2006 to 2021, revealing an escalating risk over time. Notably, the highest HQ value is observed in 2021, reaching an alarming level of 13.74, significantly surpassing the WHO's established standard of HQ=1. Simultaneously, the carcinogenic risk values also demonstrate a consistent increase throughout the mentioned years in Lahore. In conventional human health risk assessment, a cancer risk (CR) value of 1 in a million ( $10^{-6}$ ) is deemed acceptable. However, for arsenic (As), environmental policies necessitate adjusting this threshold to  $10^{-4}$  (Buchhamer et al., 2012). The progressive increase in carcinogenic risk is evident, with the highest recorded value in 2021 reaching 0.0067. These findings raise serious concerns as both the non-carcinogenic and carcinogenic effects of arsenic pose detrimental implications for the studied population.

Non-carcinogenic risks can contribute to a range of health complications, encompassing integumentary, hematopoietic, respiratory, nervous, immune, renal, and hepatic malfunctions (Kumar & Singh, 2020).

To effectively reduce human health risks, policymakers should focus on key strategies. Firstly, they should prioritize the management of pollutant discharge from various sources such as industrial wastewater and domestic sewage. Implementing stringent regulations and control measures will minimize the release of harmful substances into the environment. This includes investing in advanced treatment technologies, enforcing compliance, and promoting sustainable practices to mitigate pollution risks. Secondly, policymakers need to adopt a comprehensive approach to land management. This involves establishing groundwater protection areas to prevent further contamination from anthropogenic activities. Additionally, a risk-based land management approach should be adopted, integrating environmental land zoning into land-use planning processes. This enables the identification and mitigation of potential risks, ensuring that land-use decisions align with environmental considerations and public health goals. By prioritizing effective management of pollutant discharge and implementing protective measures for groundwater resources, policymakers can significantly reduce human health risks.

#### 4. Conclusion

The study highlighted the effects of land use and land cover changes on the concentration of Fluoride and Arsenic in the ground water of Lahore, Pakistan. Based on the variation that occurred in the quality of groundwater due to Land use and land cover changes, the possible health risk assessment was also performed. The results of the spatial variation showed that the land use and land cover pattern of the study has undergone a remarkable change, with built-up being the increasing class followed by barren land while vegetation class has been observed to be reducing with every passing year. Temporal results showed that the concentration of Fluoride and Arsenic has been observed to be increasing from 2006 to 2021. Arsenic showed a distinct increasing pattern in the built-up part of the land cover of Lahore followed by barren land while Fluoride did not show any distinct pattern in terms of land use and land cover class like Arsenic, but an increasing concentration in each of the land use and land cover class has been seen throughout the passing years. The increasing concentrations are attributed to both geogenic as well as anthropogenic activities including unorganized use of land, majorly in terms of industrialization and rapid urbanization.

Because of the increasing concentration of Fluoride and Arsenic in the groundwater of Lahore, the residents are exposed to a number of health issues. The health risk assessment done in this study showed increasing risk of both carcinogenic and non-carcinogenic risks. The risk has been seen to be increasing from the year 2011 to 2021 for both Fluoride and Arsenic. To overcome this issue, proper policies related to reasonable use of land should be framed and implanted strongly. This will help in reducing the excessive land use land cover changes and will thus reduce groundwater contamination. Consequently, the associated health risks will also be controlled in this way.

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