“Evaluation, Health Impacts & Mitigation of Heavy Metals in Ambient Air of Dehradun City”

A Thesis Submitted to the
University of Petroleum and Energy Studies

For the award of
Doctor of Philosophy
in
Health, Safety and Environment

By
ABHINAV SRIVASTAVA

September 2021

Supervisor
Dr. N.A.SIDDIQUI

Department of HSE & Civil Engineering
School of Engineering
University of Petroleum and Energy Studies
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DECLARATION

I hereby declare that this submission is my own work and that, to the best of my knowledge and belief, it contains no material previously published or written by another person nor material which has been accepted for the award of any other degree or diploma of the university or other institute of higher learning, except where due acknowledgment has been made in the text.

ABHINAV SRIVASTAVA
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DATE- 08/09/2021

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THESIS COMPLETION CERTIFICATE

This is to certify that the thesis entitled ***"Evaluation, Health Impacts & Mitigation of Heavy Metals in Ambient Air of Dehradun City"*** submitted by **Mr. Abhinav Srivastava** to *University of Petroleum and Energy Studies* for the award of the degree of *Doctor of Philosophy* is a bonafide record of the research work carried out by him under my supervision and guidance. The content of the thesis, in full or parts have not been submitted to any other Institute or University for the award of any other degree or diploma.

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ABSTRACT

Metals are found in nature at different levels and some are essential elements which are required as major constituents. But certain metals and the high concentration of metals can have detrimental effect on the environment and human health. Human activity affects the geological and biological redistribution of metals through pollution of the air, water, and soil. Similarly, heavy metals in the environment may be added up in the environment from different activities and they may enter into the environment by a wide range of processes and pathways. Atmospheric emissions of these metals are probably the most harmful to the environment, and consequently, to human health.

The major hazardous metals of concern for Dehradun city in terms of their environmental load and health effects are lead, mercury, chromium, cadmium, copper etc. The source of the pollution is mostly anthropogenic- industrial activity, vehicles, etc. Natural causes like seepage from rocks, and forest fires can also contribute. Generally heavy metal toxicity can cause chronic degenerative diseases the symptoms being mental disorders, pain in muscle and joints, gastro intestinal disorders, vision problems, chronic fatigue, and susceptibility to fungal infections. Industrial workers and populations living near the polluting industry are more susceptible and have to be monitored.

In the present work, different locations of Dehradun city like Sahaspur, ISBT, Ballupur, Doon hospital road, Dalanwala, Ghantaghar, Premnagar, Jakhan, Selaqui Industrial area have been chosen for the study for knowing the concentration of the eight heavy metals in these areas. From the study it was concluded that few areas like Ghantaghar, Premnagar and Selaqui Industrial area are having high concentration of some of the heavy metals that have direct correlation with different human chronic diseases. Hence, management priority should be to mitigate the problem. Physico-chemical as well as bioremediation solutions are being tried to reduce the environment load, preferably at the site of generation. While large

industries should be forced to set-up their own effluent treatment plants, common effluent treatment facilities can be considered for smaller industries, provided they are maintained. Finally, a list of specific recommendations for mitigating the problem discussed so that these areas should start implementing the methods early before the concentration of metals increase at alarming rate.

ACKNOWLEDGEMENT

It is not possible to prepare anything without the assistance and encouragement of people. This one is certainly no exception.

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Abhinav Srivastava

Date- 08/09/2021

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ABBREVIATIONS

Acronym	Full Form
%	Percentage
°	Degree
°C	Degree Celsius
$\mu g/m^3$	Microgram Per Cubic Metre
AAP	Ambient Air Pollution
AAS	Atomic Absorption Spectrophotometer
As	Arsenic
Br	Bromine
Cd	Cadmium
CO_2	Carbon Dioxide
CO_3	Carbon Trioxide
CPCB	Central Pollution Control Board
Cr	Chromium
Cu	Copper
DNA	Deoxyribonucleic Acid
ESP	Electrostatic Precipitators
Fe	Iron
FF	Fabric Fliters
g/cm^3	Gram Per Cubic Centimetre
GIS	Geographic Information System
HC	Hydrocarbons
HCl	Hydrochloric Acid
Hg	Mercury

HNO_3	Nitric Acid
HVS	High Volume Samplers
ICP-MS	Inductively Coupled Plasma Mass Spectroscopy
kg ha^{-1} yr^{-1}	Kilogram Per Hectare Per Year
mg/m^3	Milligram Per Cubic Metre
$mgkg^{-1}$	Milligram Per Kilogram
mL	Milli Litre
Mm	Millimetre
Mn	Manganese
ng/m^3	Nanogram Per Cubic Metre
Ni	Nickel
Nm	Nanometre
NOx	Oxides Of Nitrogen
O_3	Ozone
OSHA	Occupational Safety And Health Administration
Pb	Lead
PM	Particulate Mater
Ppm	Parts Per Million
PTFE	Polytetrafluoroethylene
RSPM	Respirable Suspended Particulate Matter
SEM-EDX	Scanning Electron Microscopy-Energy Dispersive X-Ray Analyses
SO_2	Sulphur Dioxide
SPM	Suspended Particulate Matter
SRMS	Standard Reference Materials

TSP	Total Suspended Particulate
USEPA	U.S. Environmental Protection Agency
V	Vanadium
VOCs	Volatile Organic Compounds
WHO	World Health Organization
XRF	X-Ray Fluorescence Analysis
Zn	Zinc
Mm	Micrometre

CHAPTER-1

INTRODUCTION

CHAPTER 1

INTRODUCTION

Background of the air pollution:

According to the World Health Organization, air pollution is the contamination of the environment both indoor and outdoor by any type of chemical, physical or biological agents that may lead to modification in the natural characteristics of the atmosphere (WHO, 2011). The U.S. Environmental protection agency (US EPA) defined the air pollution as a mixture of solid particles and gases in the air (US EPA, 2013a). Metals occur naturally in the environment and they do not degrade. They are found in soils and rocks, and thus any activity which disturbs the soil, such as mining or erosion, can release these metals into the air as particulate matter. Anthropogenic sources of metals include mineral processing industries, power stations using fossil fuels, refineries, brickworks and motor vehicles. Combustion generated particles are generally smaller than geologically produced particles.

There is increase in various developmental activities in different sectors such as industrial, transportation and other related fields in India and the rising problem of air pollution is catching attention of the policymakers, stakeholders and common man as well. While, different stringent steps have already been taken in some megacities of India to deal with the air pollution problem, many small cities and towns are still exposed this problem. Recent epidemiological studies have shown that total suspended particulate (TSP) matter considerably influences respiratory health. Association between suspended particulate matter and lung function parameters, respiratory symptoms and mortality have also been found. Suspended particulate matter (SPM) present in the polluted air can be absorbed into human lung tissues during breathing and cause respiratory and cardiovascular problems. Medical data suggest that it is the particulate matter that become deeply imbedded in human lung tissue and causes respiratory problems and exacerbates them and other cardiovascular diseases. In addition to negative health effects, particulate matter reduces visibility and accelerates the deterioration of buildings.

Chemical composition of particulates, especially in terms of heavy metals content, is a matter of concern due to the both acute and chronic adverse health effects linked with heavy metals. Heavy metals generally have specific density more than 5 g/cm^3. The main threats to human health from heavy metals are associated with exposure to lead, cadmium, mercury and arsenic (arsenic is a metalloid, but is usually classified as a heavy metal). Long-time exposure to toxic trace metals such as arsenic, cadmium, chromium, lead even at low concentrations can causes cancer.

We have focused our study on eight heavy metals that includes Copper (Cu), Nickel (Ni), Lead (Pb), Cadmium (Cd), Arsenic (As), Chromium (Cr), Iron (Fe) and Mercury (Hg) which are released in large amount of metric tons per year in ambient air.

Sources of Heavy metals:

Natural source:

Activities like volcanic eruption, jungle fires, cyclonic disturbances and cosmic dust are contaminating the ambient air through the release of rare earth metals like Pb, As, Cd and Cr in the form of particulate matter. While the maximum dispersion of heavy metals i.e. 80% is accounted from the volcanic activity and the remaining 20% is released from other biogenic sources and the forest fires.

Anthropogenic source:

Human interference with the environment has increased tremendously. Major sources of heavy metal emissions in the atmosphere are mining, energy production, construction, non-renewable fuel combustion, municipal waste combustion, vehicular movement, construction material production and demolition activities. In mining industries, due to high-temperature refining process, metals such as Cu, Cr, Ni are dispersed. The coal contains different elements in trace quantity and thus the transportation of coal and coal mine fire releases particulate matter in which the heavy metals are integrated.

In cement plants and phosphate fertilizer complexes, raw materials, fossil fuels and waste fuels could cause significant emission of heavy metals, such as Pb and Cd. Heavy metals can also be added in the environment from different sources other than the industries. For example, Pb, Cu, and Zn are related to the abrasion of mobile part of motor vehicles (brakes, tires). Likewise, tire rubber is rich in Zn as well as in Cu, Pb, Mn, Co, Ni and Cd, and antimony (Sb) is identified as coming almost exclusively from the excessive use of the vehicle brakes (Gietl et al., 2010). Tires used as combustible waste in industries kilns to supplement traditional carbonaceous fuels are also a good source of the cited heavy metals. The main source of Ni and Vanadium (V) are the burning fossil fuels and oils (Al-Momani et al., 2005; Pacyna, 1984). Lead, used to be added to petrol, was the source of high levels of lead in the air of major cities. As a result of EPA‘s regulatory efforts to remove lead from on-road motor vehicle gasoline, emissions of lead from the transportation sector dramatically declined by 95% between 1980 and 1999, and levels of lead in the air decreased by 94% between 1980 and 1999. The major sources of Pb emissions to the air today are ores and metals processing and piston-engine aircraft operating on lead aviation gasoline. Arsenic gets added through several means including industrial sources, such as smelting and microelectronic industries. Cadmium gets added through incineration of municipal waste. Environmental pollution by chromium has been mostly by the hexavalent chromium in recent years. Figure 1.1 depicts the percentage of heavy metals in the environment and it’s possible sources.

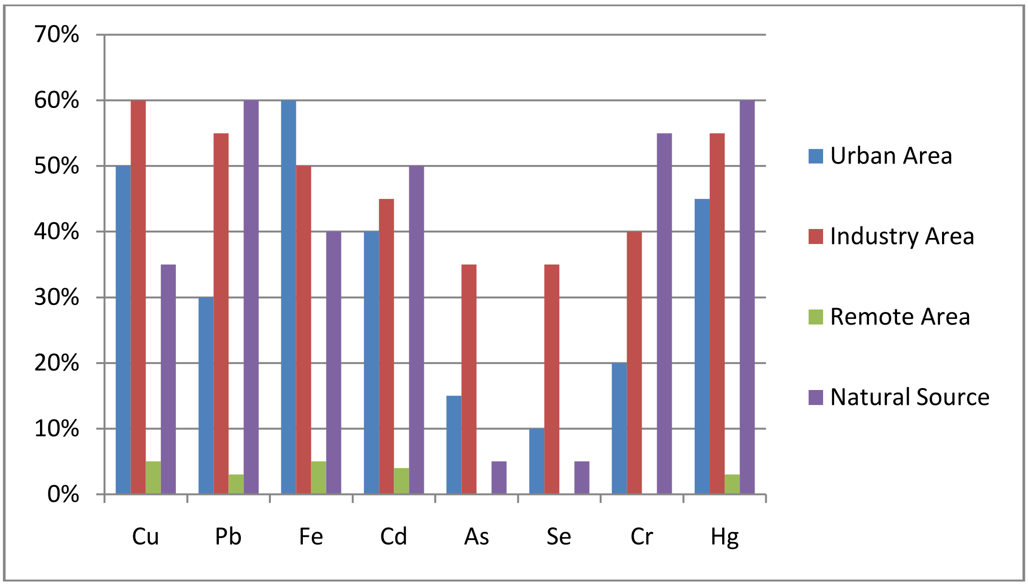

Figure 1. 1 Graph representing the heavy metals pollutant possible Sources

Heavy metals presence in air and their impact on health:

Humans may directly get in contact with heavy metals by consuming contaminated food stuffs, sea animals, drinking of water, and through inhalation of polluted air as dust fumes, or through occupational exposure at workplace (Ming-Ho et al., 2005). It has been found in several reports that the major threats on human health are due to exposure of lead, cadmium, mercury and arsenic. These metals have been studied extensively and the effects of it on human health are regularly reviewed by the World Health Organization (WHO). Heavy metals have been used by humans for several years and their adverse health effects have been known earlier also.

Heavy metal toxicity can lead to several health effects in the body. Heavy metals can damage and alter the functioning of organs such as the brain, kidney, lungs, liver, and blood. Heavy metal toxicity can either be acute or chronic effects. Long-term exposure of the body to heavy metal can progressively lead to muscular, physical and neurological degenerative processes that are similar to diseases such as Parkinson‘s disease, multiple sclerosis, muscular dystrophy and Alzheimer‘s disease. Also, chronic long-term exposure of some heavy metals may cause cancer (Jarup et al., 2003).

It has been observed that the biggest polluting countries are typically those which possess industries with huge production capabilities and the large population. The top 10 polluted countries in the world are China, the United States of America, Russia, India, Japan, Germany, Canada, the United Kingdom, South Korea and Iran (AFOP, 2014). In order to, safeguard our planet from environmental catastrophe these countries need to work towards implementing new regulations and control of their carbon dioxide emissions. This means reducing energy use and providing more forms of sustainable energy, improving transportation networks by offering improved public transport and looking for alternative fuels. For the cleanest cities, 32 cities reported a $PM_{2.5}$ reading of less than 5. Cities with the lowest level of pollution were located in Canada, the United States, Finland, Iceland and Sweden (Park, 2014). Particulate matter (PM) toxicity occurs primarily by fine and ultrafine particles due to their physical and chemical characteristics. Although, the exposure to the air pollution and especially, to fine $PM_{2.5}$ is defined as carcinogenic to humans and are toxic to their health although the mechanism pathways are not yet fully identified.

As per WHO, air pollution have been the cause of death for about 7 million people in 2012, making it the world‘s single biggest environmental health risk. In 2014, the report of the Ambient Air Pollution (AAP) data 24 base contains results of outdoor air pollution monitoring from almost 1600 cities in 91 countries (WHO, 2014d). WHO published a recent study mentioning that Delhi is the most polluted city in the world, with a $PM_{2.5}$ concentration of 153 $\mu g/m^3$ and PM_{10} concentration of 286 $\mu g/m^3$ which is extremely higher than the permissible limit as given in Table 1.1. The data points out that 13 of the 20 most polluted cities are in India including New Delhi, Patna, Gwalior and Raipur on the top four. Other cities with high levels of pollutants were located in Pakistan and Bangladesh (Park, 2014). Similarly, Beijing‘s government notified that their $PM_{2.5}$ concentrations stood at a daily average of 89.5 μg/m^3 in 2013, which is 156% higher than the national standard set for it awarding Beijing 17th rank in the WHO database (WHO, 2014e). The limits of PM set by WHO, the United States-Environmental Protection Agency (USEPA) and Central Pollution Control Board (CPCB) are summarized in Table 1.1.

Table 1. 1 Values recommended by WHO, USEPA and CPCB for atmospheric particulate matter (PM).

Particles	Exposure time	WHO (2018) (μg/m^3)	USEPA (2020) (μg/m^3)	CPCB (2009) (μg/m^3)
PM_{10}	24-hour mean	50	150	100
	Annual mean	20	-	60
$PM_{2.5}$	24-hour mean	25	35	60
	Annual mean	10	12	40

Heavy metal-induce oxidative stress and oxidation of biological molecules:

Certain heavy metals are known to generate free radicals which may lead to oxidative stress and cause other cellular damages. The mechanism of free radical generation is dependent on the specific type of heavy metal (Valko et al., 2005).

The permissible limit of heavy metals and their adverse effects on health are tabulated in Table 1.2.

Heavy metal-induced carcinogenesis:

Some heavy metals are known to have carcinogenic effect. Several signaling proteins or cellular regulatory proteins that participate in apoptosis, cell cycle regulation, DNA repair, DNA methylation, cell growth and differentiation are targets of heavy metals (Kim et al., 2015). Thus, heavy metals may induce carcinogenic effect by targeting a number of these proteins. More so, the carcinogenic effects of certain heavy metals have related to the activation of redox-sensitive transcription factors such as AP-1, NF-κB and p53 through the recycling of electrons by antioxidant network. These transcription factors control the expression of protective genes that induce apoptosis, arrest the proliferation of damaged cells, repair damaged DNA and power the immune system (Valko et al., 2005). Metal signalization of transcription factor AP-1 and NF-κB has been observed in the mitogen-activated protein (MAP) kinase pathways where the nuclear transcription factor NF-κB, is involved in controlling inflammatory responses while AP-1 is involved in cell growth and differentiation (Valko et al., 2005).The p53 protein is an important protein in cell division as it guards a cell-cycle checkpoint and control cell division (Chen, 2015). Inactivation of p53 allows uncontrolled cell division and thus p53 gene disruption has been associated with most human cancers. Also, AP-1 and NF-κB family of transcription factors are involved in both cell proliferation and apoptosis, and also regulate p53. Heavy metals generated free radicals inside the cell selectively activates these transcription factors and thus, may suggest that cell proliferation or cell death may be related to the exposure to carcinogenic metals. There exist various mechanisms of heavy metal-induced carcinogenesis.

Heavy metal-induced neurotoxicity:

Some heavy metals such as lead and manganese may affect the brain and cause neurological toxicity (Neal and Guilarte, 2012).

Table 1. 2 Permissible exposure limit for heavy metals in industrial and non-industrial areas and their adverse health effects

Sr. No	*Metals*	*Permissible limits as per WHO, OSHA, CPCB*	*Exposure effects on human*
1.	Arsenic (As)	0.6 ng/m^3 (CPCB) 10 mg/m^3 (OSHA) 1 µg/m^3 (WHO)	Gastrointestinal effects (Nausea, diarrhea, abdominal pain) central and peripheral nervous system disorder and lung cancer.
2.	Cadmium (Cd)	5 ng/ m^3 (WHO) 5 mg/m^3 (OSHA)	Cadmium build-up in the kidneys,effects on the liver, lungs, bones & immune system.
3.	Chromium (Cr)	1 µg/m^3 (WHO) 1 mg/m^3 (OSHA)	Shortness of breath, coughing, wheezing and other effects on the respiratory tract.
4.	Lead (Pb)	1 µg/m^3 (CPCB) 0.15 mg/m^3 (OSHA)	CNS, blood pressure, kidneys and vitamin D metabolism.
5.	Mercury (Hg)	0.3 µg/m^3 (WHO) 0.1 mg/m^3 (OSHA)	Kidney damage; Alteration in testicular tissue, increased resorption rates, possible human carcinogen.
6.	Selenium (Se)	0.2 to 30 ng/m^3 (WHO) 0.2 mg/m^3 (OSHA)	Loss of hairs, excessive tooth decay and discoloration, garlic odour in breath and urine, lack of mental alertness.
7.	Copper (Cu)	1 mg/m^3 (OSHA)	Gastrointestinal bleeding, liver, kidney failure and death.
8.	Iron (Fe)	10 µg/m^3 (USEPA)	Gastrointestinal mucosa, stomach and intestinal erosions and ulceration.

Penetration, deposition and clearance of airborne particles:

The main route of entry for ambient PM to the body is *via* inhalation. In general, the particle deposition in the human respiratory tract is determined by biological factors, such as lung morphology and breathing patterns, physical factors, such as fluid dynamics, particle properties (shape, charge, density, hygroscopicity and size), and deposition mechanisms (Hofmann, 2011). These factors determine three deposit areas: the nasal-pharyngeal, trachea-bronchial and alveolar regions. Depending on the particle size and mass, the deposition rate varies between these regions. Particles larger than 3µm and smaller than 0.003µm tend to deposit in the nasal-pharyngeal region. $PM_{0.1}$ is distributed throughout the respiratory tract and act as gases leading to an increase in the deposition rate at the extra-thoracic level (Patton and Byron, 2007). Particles between 0.003µm and 3µm reach the alveolar region, with a maximum deposition for 20-30 nm particles. About 50% of $PM_{0.1-2.5}$ is able to reach the alveoli, carrying along the adsorbed potentially toxic compounds.

Particle deposition in the respiratory tract can be made by five mechanisms: inertial impaction, sedimentation, diffusion, interception and electrostatic interaction (Carvalho et al., 2011). Inertial impaction takes place in the nasopharyngeal region and in the trachea-bronchial tree where the air speed is high and the flow is turbulent. This mode characterizes the large particles having a diameter bigger than 10µm. Sedimentation includes particles having a diameter between 1 and 25 µm and it occurs at the trachea-bronchial tree and the alveolar region. Diffusion, also known as Brownian motion, is correlated to particle size and it is especially possessed by the particles which have size < 0.5 µm. Interception on the other hand, takes place when the particle, without deviating from its path, meets the wall of a duct pulmonary and deposits there. Finally, the electrostatic interaction depends on the electrostatic charges of atmospheric particles and it is characteristic of particles smaller than 0.5 µm.

After deposition, the body tries to reduce the retention of inhaled particles by clearing them from the lung. The term ―clearance‖ was introduced to describe the translocation, transformation and removal of deposited particles from the various regions of the respiratory tract. In the upper respiratory tract mainly PM_{10} are deposited and in this part of the lung the clearance is performed by coughing. $PM_{2.5-10}$ get deposited onto ciliated surfaces and they are removed by the ―mucociliary escalator‖ (rapid elimination in 24 hours). The fine and ultrafine fractions of PM ($PM_{2.5}$) mainly deposit on the alveolar region. After deposition, the particles could be phagocytosed by macrophages and get transported to upper lung regions for mucociliary clearance or the particle-loaded phagocytes enter the lymphatic system (slow clearance). Depending on the inhaled concentrations of particles, a considerable part of the particles are taken up by alveolar epithelial cells, which act in addition to macrophages as initiators of inflammatory responses. Ultrafine particles taken up by epithelial cells are considered to escape from the clearance done by alveolar macrophages. A fraction of these particles is able to reach the systemic circulation (Nemmar et al., 2002; Oberdörster et al., 2005; Semmler-Behnke et al., 2007).

Local context; Dehradun city:

Dehradun is the capital city of the state Uttarakhand which lies in the Doon Valley, in the foothills of the Himalayas between the river Ganges on the east and the river Yamuna on the west, with an estimated urban population of 1.2 million. Dehradun is famous for its picturesque landscape and beautiful weather that provides a gateway to the surrounding region. The place is largely known famous for its Basmati rice, Litchi, tea gardens, Sal forests and water canals. All these natural bodies are deteriorating at high speed due to extensive unplanned development and expansion.

Geographic, demographic and meteorological data:

Dehradun coordinates are 30 degree 19' North and 78 degree 03' East. Dehradun district can be seen on the North West corner of the state, located between the latitudes 29

°58‘ N and 31°2‘ N and longitudes 77° 34‘ E and 78° 18‘E. It is located on the foothills of the Himalayas and its border by the Himalayan range to the north, the Shivaliks to the south, the Ganges to the east and the Yamuna River to the west. The lowest point is Raiwala at 315 meters above sea level, and the Tuini hills is the highest point, rising to 3700 meters above mean sea level. The famous hill stations, Mussoorie and Chakrata, are part of the district and lie in the lesser Himalayan ranges. The district on the north and the North West is bounded by the district of Uttarkashi and Tehri and Pauri districts are on the east. In the West borders are Sirmaur district of Himachal Pradesh and to the south are Haridwar and Saharanpur district of Uttar Pradesh (as shown in figure 1.2). Dehradun is situated in the zone IV (High damage risk zone) as per the Earthquake hazard zoning of India.

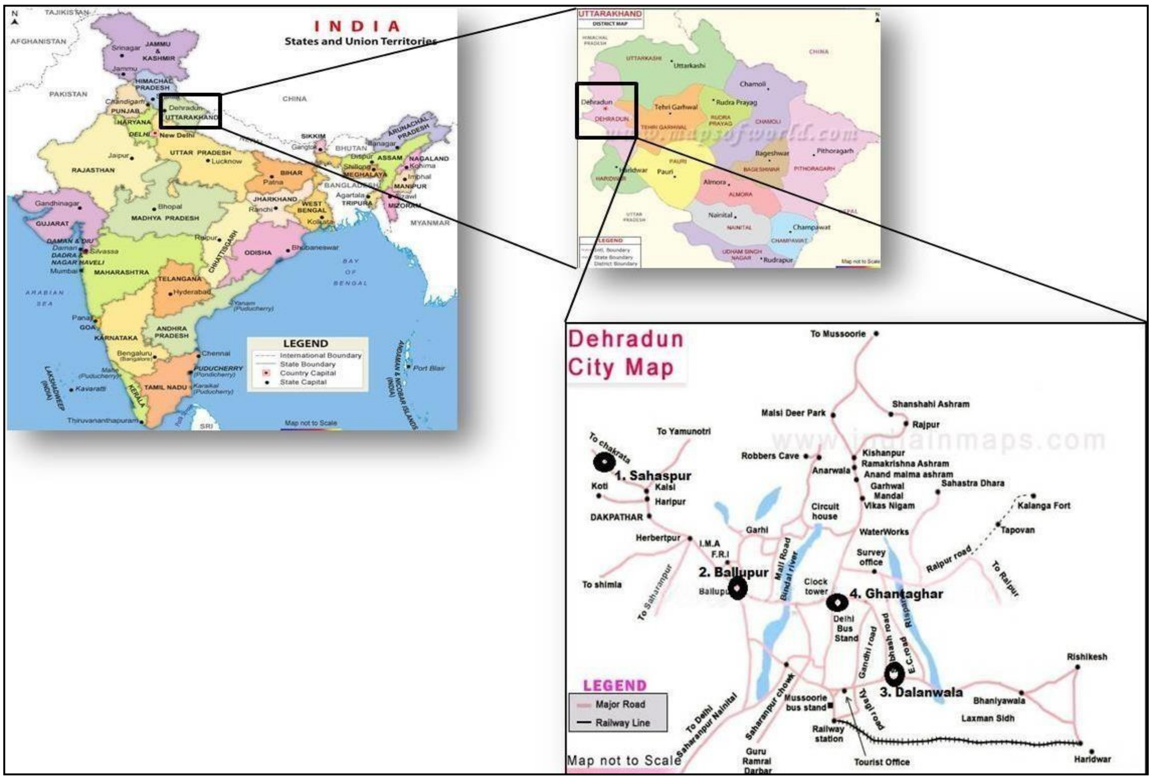

Figure 1. 2 Geographical location of Dehradun city.

Dehradun has an extreme type of continental climate due to its distance from the sea providing it with continental air during major parts of the year. It mainly receives monsoon during the three monsoon months of July, August and September and causes increased humidity, cloudiness and rain.

Most of the annual rainfall is received during months from June to September whereby July and August receiving heavy rainfall. The rainfall and temperature variations within the city are quite considerable due to altitudinal variation and local geography with presence of high Mussoorie hills. According to the India Meteorological Department classification, seasons in India are divided as Winter Season (January to February), Summer Season (March to May), Monsoon or South-West Season (June to September) and Post-monsoon Season (October-December). But, for the study of climate of Dehradun season are classified as:

1. Winter Season (December to February)
2. Summer Season (March to May)
3. Monsoon or South-West Season (June to September)
4. Post-monsoon Season (October-November).

The summer season in Dehradun is moderately hot while winters are extremely cold. The temperature may go up to 41 °C in summers and the average temperature recorded is around 35-36 °C. In winters the temperatures may fall to 1-2 °C for a few days when the cold winds from the nearby Himalayan Mountains makes the winters severe. The winters are also show the presence of mist and fog on few days especially during night and early in the mornings.

During the summer months, the temperature ranges between 36◦C and 16.7◦C. The winter months are colder with the maximum and minimum temperatures touching 23.4◦C and 5.2◦C, respectively. Dehradun experiences heavy to moderate showers during late June to mid-August. Most of the annual rainfall (about 2000 mm) is received during the months from June to September, July and August being the rainiest months of the season.

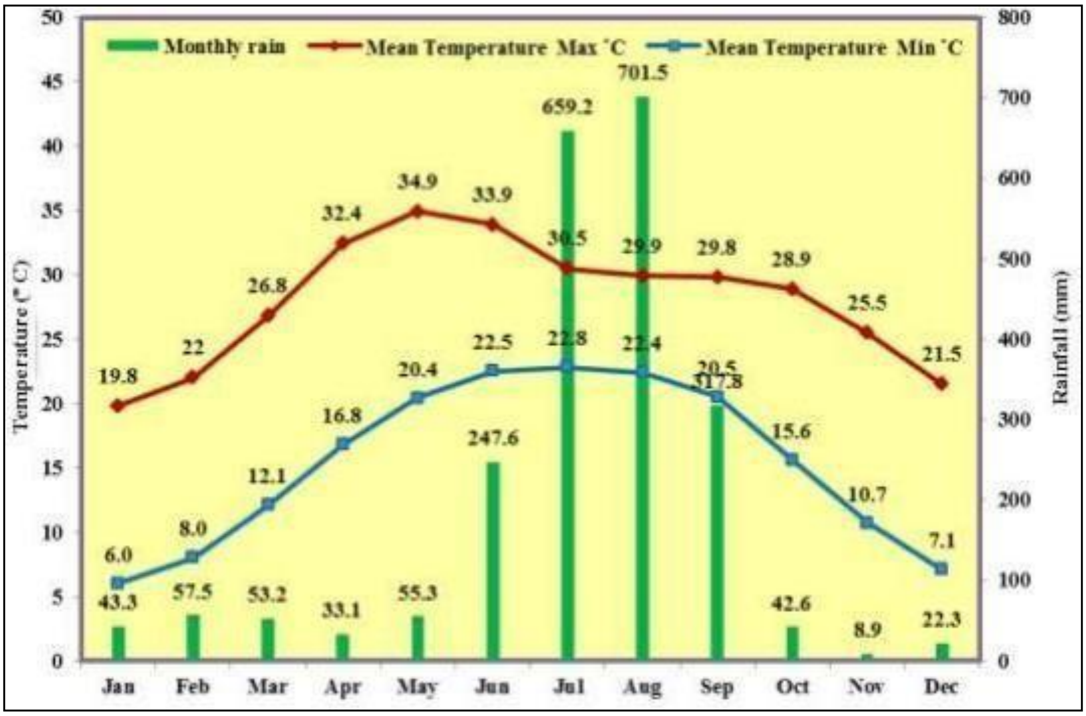

Figure 1. 3 (a) Monthly maximum and minimum temperature variation.

Reference http://amssdelhi.gov.in/news_events/Dehradun_Climate.pdf

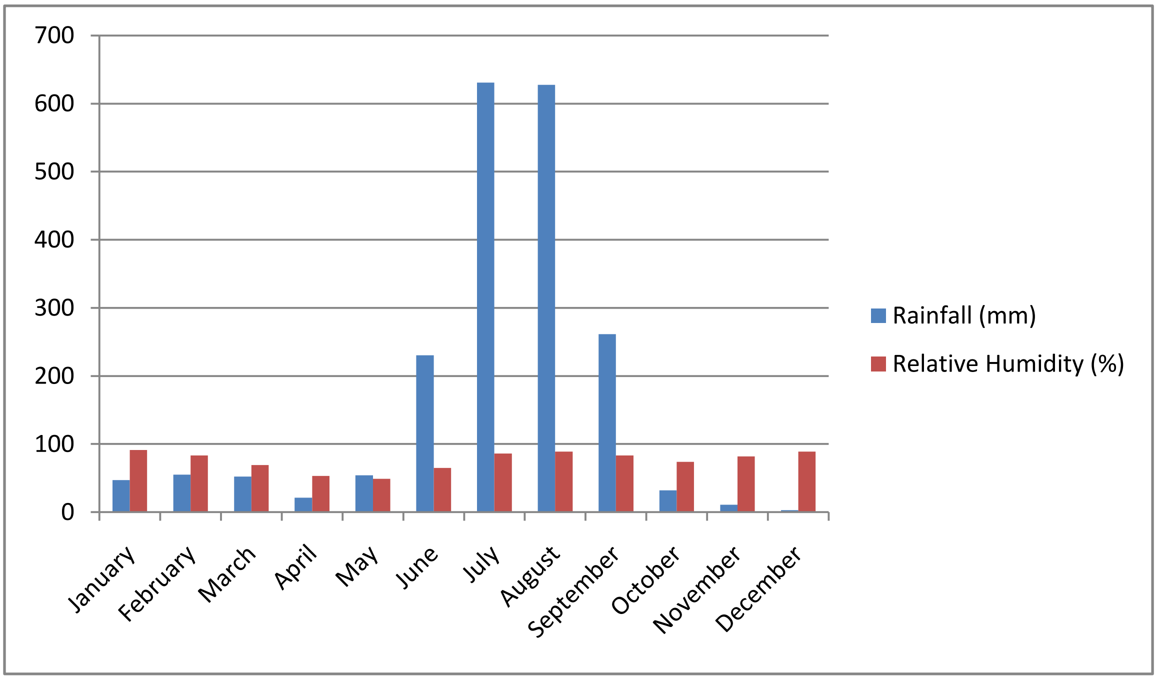

Figure 1.3 (b) Average Annual Rainfall and Relative Humidity of Dehardun

Reference: http://dehradun.nic.in/climate/

Commercial importance:

The Dehradun city has emerged as an important business, educational and cultural destination in North India after becoming the capital of newly carved out Uttarakhand state since the year 2000. Moreover, city is the wholesale trading center for the entire hill region of the Uttarakhand state. The city is also well known for its salubrious climate, natural beauty, places for tourist‘s attraction and institutions of national and international importance. Therefore, about one million Indian and foreign tourists visit the city every year in the form of floating population. The resident population of the city grew alarmingly during 1991–2011 and it registered a growth of about 114% during the last two decades. In addition, the population of the Dehradun city will grow at the rate of 3.5% from 2010 to 2014, and 3.0% from 2015 to 2019 (UUDP 2007). Several industries namely, pharmaceutical and chemicals, electronic and electrical engineering, food processing and glassware were established in Dehradun city after the declaration of state industrial development policy in the year 2003. This growth of population and industrialization was accompanied by alarming growth in built up area of Dehradun city.

Table 1. 3 Maximum permissible limit for heavy metals as per WHO and OSHA

S. No.	Heavy Metal	Prescribed Conc. Limit Living Standards (USEPA, WHO & CPCB)	Max Permissible Limit[2] Industrial Standards (OSHA) (
1	Lead (Pb)	1 µg/m^3 (CPCB)	0.15 mg/m^3
2	Iron (Fe)	10 µg/m^3(USEP A)	5 mg/ m^3
3	Chromium (Cr)	1 µg/m^3 (WHO)	1 mg/m^3
4	Cadmium (Cd)	5 ng/ m^3 WHO	5 mg/ m^3

Commercial built up area in the city registered a growth of about 322% during 1982–2004. The major conversion into built up area in the city took place from agricultural land, forest areas and open spaces. Alarming increase in vehicular traffic has been observed in the city after the initiation of growth in population, urbanization and industrialization process. A growth of about 362% was registered in the registration of vehicles in the last decade (UUDP 2007). Significant rise is also noticed in the number of vehicles that registered elsewhere but plying on the roads of Dehradun city and are emitting huge amount of heavier molecules, hydrocarbons, NOx, carbon trioxide (CO_3), carbon dioxide (CO_2), etc. Suspended particulate matter (SPM) concentration in the city exceeds the National Ambient Air Quality Standards. During 2003–2005, the level of SPM has increased from 250 to 400 $\mu g/m^3$ in the city (UUDP 2007).

Due to recent multiplying population growth, urbanization, industrialization and several unplanned development projects in Dehradun city have resulted in large scale ecological degradation which in turn has greatly affected the urban climate on alarming scales. Hence, in the present study we have chosen nine different sites (Sahaspur, ISBT, Ballupur, Doon hospital road, Dalanwala, Clock Tower, Premnagar, Jakhan, Selaqui Industrial area) in the city and measured the level of heavy metals in these areas to estimate the overall air quality of the Dehradun.

Scope of the work:

The motivation for this work is the body of epidemiological studies that associate elevated concentrations of ambient $PM_{2.5}$ and PM_{10} with increased human mortality and morbidity. Although the specific mechanisms of health damage that resulted in these associations had not been demonstrated, numerous toxicological studies provided considerable evidence for several plausible mechanisms. Role of air borne heavy metals could be one of them which are not studied rigorously in Dehradun till now. Hence, to contribute to fulfil this gap of knowledge and to better understand how air borne heavy metals are affecting the public health in Dehradun city this study was done.

Eight common heavy metals which are: arsenic, cadmium, chromium, lead, mercury, selenium, copper and iron are generally found in air with large amount, they can be dangerous. Generally, humans are exposed to these metals by ingestion (drinking or eating) or inhalation (breathing). Working in or living near an industrial site which utilizes these metals and their compounds increases ones risk of exposure, as does living near a site where these metals have been improperly disposed. In fact these are the mediums through which it gets into the air particles. Acute and chronic exposure to these heavy metals may leads to vomiting, diarrhoea, lung damage, fragile bones, asthma, skin ulcers, and some of them also possess carcinogenic properties. There is possibility that several cases of health issues pertinent in the regions may be due to the growing concentrations of heavy metals in air particles and which is not addressed yet because of lack of awareness and initiatives in the field.

Objectives of the study:

The principal objectives of this work are:

- To identify the major industrial and non-industrial sources responsible for the emission of heavy metals in air.
- To study and analyse different types of heavy metals present in ambient air of Dehradun city. (12 months)
- To correlate their concentrations with hazardous health effects on human.
- To observe possible mitigation measures.

---End of Chapter 1---

CHAPTER-2

REVIEW OF LITERATURE

CHAPTER 2

REVIEW OF LITERATURE

In Dehradun city, due to the increment in the construction and rapid growth of industries, the air quality is degrading. According to the WHO study, Dehradun is ranked as the 30th most polluted city in the world. Furthermore, the geographical and meteorological conditions of the city does not allow for the easy dispersal of the pollution. Therefore, the assessment of its air quality is of prime concern and in the present study, an attempt is made evaluate the presence of heavy metals in the ambient air at different sites in Dehradun and list out the affected areas which are most affected, so as to apply the major mitigation methods there and stop further deterioration of the air.

For the literature review, the complete study is divided in four sections:

1. Research studies assessing the air quality of Dehradun and its neighboring cities.
2. Studies focusing on air pollution caused by the heavy metals.
3. Studies correlating the effect of heavy metals in air on the human health.
4. Mitigation strategies employed to bring down the contamination from the air.

Large number of research papers has been reviewed to find out the research gap and some of the recent studies have been discussed in details to get the clear insight of the present status.

Research studies assessing the air quality of Dehradun and its neighboring cities:

i. ***Chauhan et al., 2010:*** This article discusses the changes in seasonal variations of air pollutants concentrations at different places of Uttarakhand such as urban, industrial, commercial and agricultural areas. PM_{10} (RSPM), oxides of nitrogen (NOx), suspended particulate matter (SPM) and sulphur dioxide (SO_2) were collected from the 4 sites in

Haridwar and Dehradun Valley, Uttarakhand. The first site was urban area, Shivalik Nagar, Haridwar. SIDCUL was the second site, one of the most industrial areas of Haridwar. The commercial centres, famous Clock Tower of Dehradun Valley, the third site. The last site was a control site, an agricultural area where pollution level was very low. These pollutants have been observed and recorded on the monthly and seasonal basis. It was found that the concentrations of the pollutants were very high in winter season as compared the summer or the monsoon. The SPM and PM_{10} levels at all selected sites (excluding Roshnabad) have exceeds the prescribed limits of CPCB.

ii. ***Awasthi et al., 2016: T***he aim of present investigation is to elucidate the persistent increase in the concentration of particulate matter and gaseous pollutants in an area rapidly developing as industrial belt in district Haridwar after development of State Industrial Development Corporation of Uttarakhand (SIDCUL) in 2002. The analysis of the increase in the level of ambient air pollutants such as suspended particulate matter, respirable suspended particulate matter and the concentration of gaseous pollutants (SO_2 and NO_X), during a period of six consecutive years (2003-2009) at Bahadarabad. The concentration of these parameters is found to increase significantly by manifold over a period of six years of measurements. The concentrations of SPM and RSPM are compared with the concentration of gaseous pollutants SO_2 and NO_X.

Parameter ($\mu g/m^3$)	SO_2	NO_X	SPM	RSPM
SO_2	1	0.97166	0.8150446	0.7677859
NO_X	-	1	0.8455824	0.8309216
SPM	-	-	1	0.8888961
RSPM	-	-	-	1

Correlation coefficient between gaseous pollutants and particulate matter using Pearson’s product-moment correlation method.

iii. ***Deep et al., 2017:*** They have studied and analyzed some of the air quality parameters of Dehradun and Rishikesh in Uttarakhand. A large number of particulate matters are generated due to such developmental schemes. They studied the growth in the level of ambient air pollutants. The particulate matters (PM_{10}), as well as the concentration of gaseous pollutants (SO_2 and NO_2), during 2015 to 2016, are studied. Monthly as well as seasonal variations of these pollutants were analyzed and the concentration was high during pre -monsoon season as compared to the other seasons. The PM_{10} concentration is found about two times higher than the prescribed national standard while SO_2 and NO_2 levels were found within the predetermined limits, fixed by the Central Pollution Control Board (CPCB) New Delhi, India.

iv. ***Deoli et al., 2018:*** In this study, the assessment of air quality of Dehradun is done to find out the level of air pollution in the city. The air quality is measured at ten different sites of the city during winters. Different parameters, such as SPM, RSPM, SOx and NOx were monitored and also the metrological parameter like temperature, humidity and rainfall were taken into account. The concentration of pollutants was high at sites like Clock Tower, Prince Chowk and Sharunpur chowk and this elevated pollutant level was mainly due to the presence of vehicular emission.

v. ***Deep et al., 2019:*** The variations in the ambient concentrations of particulate matter (SPM and PM_{10}) and gaseous pollutants (SO_2 and NO_2) at Clock tower, Rajpur road and Inter State Bus Terminal station in Dehradun city, Uttarakhand, India are analysed for the period of 2011–2014. Mean concentrations are observed to be higher during pre-monsoon season as compared to the winter and monsoon. PM_{10} and SPM concentrations with maximum values of 203 ± 23 and $429 \pm 49 \mu g\ m^{-3}$, respectively, during winter, are found to exceed the national standards by factors of 2 and 3. Winter-time elevated pollution in Dehradun is attributed to the lower ventilation coefficient (derived from Era interim model fields) and minimal precipitation. Nevertheless, the SO_2 and NO_2 levels are observed to be within the criteria notified by the CPCB, India.

Studies focusing on air pollution caused by the heavy metals:

i. ***Mishra et al. 2013:*** In this study, the ambient concentrations of PM_{10} and associated heavy metals measured at three sites in the mining areas of Jharia coalfield during October 2012-February 2013. After acid digestion, quantification of 5 major heavy metals (Co, Cd, Mn, Ni and Pb) was done using Atomic Absorption Spectrophotometer (AAS). Out of 5 metals studied two metals (Ni and Pb) are criteria pollutants as per NAAQS (2009). As per the magnitude of the mean concentrations, these can be ranked in the following order Mn > Ni > Co > Cd >Pb. PM_{10} and heavy metals loading in coal mining area was found to be almost 2 times higher than that of control site. The concentration of PM_{10} and heavy metals observed in present study were compared to those reported previously from other parts of India and the world. For coal mining areas level of these pollutants was found higher than that of most of the cities. Correlation analysis showed strong relationships between Cd–Ni, Ni–Mn, and Cd–Mn.

ii. ***Mafuyai et al. 2014:*** The concentration of respirable dust at seven sampling stations in Jos metropolitan area, was measured weekly for three consecutive months (October 2012 to December 2012). Characterization of the dust particles showed that the concentration of fine dust (<2.5 μm) ranged from 55.0 - 124.9 μgm^{-3} while the coarse dust (2.5 - 10 μm) varied from 20.0 - 124.4 μgm^{-3}. FAAS (Flame atomic absorption spectrophotometer) analysis of the respirable dust shows that the concentrations of heavy metals ranged from 0.121 - 0.832 $mgkg^{-1}$ Pb, 0.019 - 0.111 $mgkg^{-1}$Cr, 0.171 - 1.081 $mgkg^{-1}$Fe, 0.002 - 0.056 $mgkg^{-1}$ Mn, 0.002 - 0.438 $mgkg^{-1}$Cd, 0.696 - 1.712 $mgkg^{-1}$Zn, 0.025 - 0.571 $mgkg^{-1}$Cu, and 0.021 - 0. 478 mg kg^{-1} Ni across the sites studied. The main sources of these heavy metals in the sampled area could be attributed to anthropogenic activities like open incineration of waste and vehicular traffic. The concentrations of Cd, Mn and Ni were found to be far above the standard limits prescribed by the WHO for respirable dust. These results convey the health risk the inhabitants in the study area are exposed to. We therefore recommend that measures be taken to regulate these anthropogenic activities.

iii. ***Pal et al. 2014:*** Samples of suspended particulate matter (PM_{10}) were collected from three different sites in Moradabad, India. The sampling was done concurrently twice a week during the period of April 2011-March 2012. Elemental concentration of PM_{10} was analyzed using an Inductively Coupled Plasma Optical Emission Spectrophotometer (ICP-OES). The monthly mean concentration of PM_{10} (RSPM) ranged between 63-226 μgm^{-3}, which was higher than the permissible limit of 100 μgm^{-3} of National Ambient Air Quality Standards. The maximum concentration of Zn, Fe, Cu, Cr and Ni found in the Industrial area of the city was 21.24, 18.43, 15.23, 0.41, 0.03 μgm^{-3}, respectively; whereas the maximum concentration of Pb (2.72 μgm^{-3}) and Cd (0.20 μgm^{-3}) was found in heavy density traffic area, denoted as commercial area. The study shows that high number of vehicles and the brassware industries are the major cause of the high concentration of heavy metals in the city.

iv. ***Khanna et al. 2015:*** The study points to the concentrations of 25 heavy metals adsorbed to fine fraction of particulate matter, $PM_{2.5}$ have been studied and analysis was done at site located on the National Highway of the capital city, Delhi. The $PM_{2.5}$ collected in winter season on the filter papers and that was analyzed for different heavy metal concentrations using ED-XRF. It was observed that the metals concentrations were in the trend: Si > K > S > Ca = Fe > Zn = Pb > Br. The potential health risks of different carcinogenic heavy metals have also been calculated.

v. ***Zheng et al. 2015:*** Airborne dust particles containing high levels of toxic metals, is recognized as one of the most harmful environment component. The purpose of this study was to evaluate heavy metals pollution in dust fall from bus stations in Beijing, and to perform a risk assessment analysis for adult passengers. The concentrations of Cd, Co, Cr, Cu, Mo, Ni, Pb, V and Zn were analysed by inductively coupled plasma mass spectroscopy (ICP-MS). The spatial distribution was determined by Geographic Information System (GIS) mapping technology. The results indicate that dust samples have elevated metal concentrations, especially for Cd, Cu, Pb and Zn. The nine metals can be divided into two categories in terms of spatial distribution and pollution level. Cd, Cr, Cu, Mo, Pb and Zn reach contaminated level and have similar spatial patterns with hotspots distributed within the Fifth Ring Road.

vi. ***Melaku et al. 2008:*** In this study, the researchers observed high seasonal variability in both ambient air and wet deposition samples. Ambient air samples were collected from Washington DC single site for 7 months. Arsenic, Chromiun, Lead and cadmium were being analyzed in the study by using acid- digestion method. The ranges of heavy metal concentrations for the 6-day ambient air samples were 0.800−15.7 ng/m3, 1.50-30.0 ng/m3, 16.8-112 ng/m3 and 2.90-137 ng/m3 for arsenic, cadmium, chromium and lead, respectively, with a precision better than 10%. The spread in the heavy metal concentration over the observation period suggests a high seasonal variability for heavy metal content in both ambient air and wet deposition samples.

vii. ***Talbi et al. 2018***: In the study, the researchers investigated the two different sites urban and roadside at Algiers (Algeria) where they measured the concentrations of PM less than 1µm, 2.5µm, 10µm and contents of heavy metals. Sampling procedure was done for two years using a high volume samplers (HVS) equipped with a cascade impactor at four levels stage. Then the characterization of the heavy metals those were associated with the PM was performed using X-Ray Fluorescence analysis (XRF). The PM_1, $PM_{2.5}$ and PM_{10} concentrations on roadside fluctuates between 13.46 - 25.59 $\mu g/m^3$, 20.82–49.85 $\mu g/m^3$ and 45.90–77.23 $\mu g/m^3$, respectively. However, at the urban site, the PM_1, $PM_{2.5}$ and PM_{10} concentrations varied from 10.45-26.24 $\mu g/m^3$, 18.53–47.58 $\mu g/m^3$ and 43.8–91.62 $\mu g/m^3$.

The heavy metals associated with the PM were confirmed by Scanning Electron Microscopy-Energy Dispersive X-Ray analyses (SEM-EDX). The different spots of $PM_{2.5}$ analysis by SEM-EDX shows the presence of nineteen elements and lead was found with maximum of 5% (weight percent). In order to determine the source contributions of PM levels at the two sampling sites sampling, principal compound analysis (PCA) was applied to the collected data. Statistical analysis confirmed anthropogenic source with traffic being a significant source and high contribution of natural emissions.

Studies correlating the effect of heavy metals in air on the human health:

i. ***Fortoul et al. 2015:*** In this review, authors have specifically focused their attention on the metals in PM and its association with the deleterious effect on human health. It has been proven that heavy metals for example cadmium, lead and mercury are major air pollutants which are added in the environment by industrial activities, combustion, and human activities. In addition, they share the possibility to exert toxic effects by the production of reactive oxygen species. The chemical nature of the inhaled metals, its exposure, route, and physiology of the organ determines the metabolism and its effects. Metals can also interact directly with the components of the cell, and get accumulated in the lysosomes, the mitochondria and lead to cell death.

ii. ***Sharma et al. 2017:*** The environment has the hazardous chemicals due to the number of natural and anthropogenic activities that are responsible for the adverse effects on the health of human and the environment. Fossil fuel combustion is held responsible for the changes in the composition of the atmospheric. Air pollutants, for example carbon monoxide (CO), ozone (O3), nitrogen oxides (NOx), sulfur dioxide (SO2), volatile organic compounds (VOCs), heavy metals, and respirable particulate matter ($PM_{2.5}$ and PM_{10}), all are very different in their chemical composition, emission, time of disintegration and diffusion ability in the environment. The damages can be from minor changes in respiratory tract to chronic respiratory problem, lung cancer, and acute respiratory infections in children and in adults. Moreover, short/long-term exposures also have been linked with premature mortality and reduced life expectancy.

iii. ***WHO, 2018:*** A major risk factor for cardiovascular disease, chronic obstructive pulmonary disease and lung cancer among adults and increased risk of acute lower respiratory infections and associated mortality among young children is caused due to indoor exposure to pollutants from the household combustion of solid fuels on open fires or traditional stoves in developing countries like India. As per WHO estimation some 80% of outdoor air pollution-related premature deaths were due to ischemic heart disease and strokes. To reduce the number of Indian automobiles running on diesel several steps has been taken and the country‘s National Green Tribunal also announced to ban more than 15 years older vehicle from the roads of New Delhi.

Mitigation strategies employed to bring down the contamination from the air:

i. ***Yang et al. 2005:*** In this research work, researchers have proposed tree planting by the municipal government as a measure to alleviate air pollution in Beijing. It is based on the analyses of satellite images and field surveys to establish the characteristics of current urban forest in the central part of Beijing. The influence of the urban forest on air quality was studied using the Urban Forest Effects Model. The results show that there are 2.4 million trees in the central part of Beijing. The diameter distribution of the trees is skewed toward small diameters. The urban forest is dominated by a few species. The condition of trees in the central part of Beijing is not ideal; about 29% of trees were classified as being in poor condition. The trees in the central part of Beijing removed 1261.4 tons of pollutants from the air in 2002. The air pollutant that was most reduced was PM_{10}, the reduction amounted to 772 tons. The carbon dioxide stored in biomass form by the urban forest amounted to about 0.2 million tons.

ii. ***Yang et al. 2008:*** In this research, the authors have checked the level of air pollution removal by green roofs in Chicago was quantified using a dry deposition model. The result showed that a total of 1675 kg of air pollutants was removed by 19.8 ha of green roofs in one year with O_3 accounting for 52% of the total, NO_2 (27%), PM_{10} (14%), and SO_2 (7%). The highest level of air pollution removal occurred in May and the lowest in February. The annual removal per hectare of green roof was 85 kg ha^{-1} yr^{-1}. The amount of pollutants removed would increase to 2046.89 metric tons if all rooftops in Chicago were covered with intensive green roofs.

iii. ***Rowe, 2011:*** Herein, the author has suggested green roofs that involve growing vegetation on rooftops and are one tool that can help mitigate the negative effects of pollution. The discussion will focus on how green roofs influence air pollution, carbon dioxide emissions, carbon sequestration, longevity of roofing membranes that result in fewer roofing materials in landfills, water quality of stormwater runoff, and noise pollution. Suggestions for future directions for research include plant selection, development of improved growing substrates, urban rooftop agriculture, water quality of runoff, supplemental irrigation, the use of grey water, air pollution, carbon sequestration, effects on human health, combining green roofs with complementary related technologies, and economics and policy issues.

iv. ***Bhandarkar, 2013:*** In this research, the author has focused on emission from vehicles especially automobiles that is the main cause for about two third of air pollution in the urban area. The major pollutants emitted by motor vehicles including CO, NOx, sulphur oxides (SO), hyrdocarbons, lead and suspended particulate matter (SPM), have damaging effects on both human health and ecology. The internal combustion engines need a mixture of air and fuel to burn and produce energy to propel the vehicle. These burnt gases which come out of the exhaust are responsible for pollution. In petrol engines, a mixture of exhaust gases includes hydrocarbons, CO and NOx. If these gases are in excess quantities, vehicular pollution is caused. Emissions of diesel vehicles are the concentration of CO and

HC in the diesel exhaust are rather low, both of which are compensated by high concentration of NOx and CO_2. There are smoke particles and oxygenated HC, including aldehydes and odour producing compounds.

v. ***Baldauf and Nowak, 2014:*** In this paper, the authors discuss about the conventional air quality management programs to reduce or eliminate air pollution emissions from the source, other options exist for planners and developers to further reduce an urban population‘s exposure to harmful concentrations of air pollutants. Given the significant impact of vehicular traffic on air pollution in most urban areas of the world, this section emphasizes and provides examples of opportunities to mitigate impacts from vehicular sources in particular. These options include the preservation and planting of vegetation, the development of roadside or near-source structures, and the modification of terrain surrounding roadways or other pollutant emission sources.

Inferences drawn from the literature survey:

From the review of literature, it was found that the analysis of air quality of Dehradun city is an important concern that still has not been addressed properly by the researchers and needs intervention. Lots of construction work and the immense growth of population with increasing industries are imposing serious impact on the environmental air quality. Unhealthy atmospheric conditions in the city center and other neighboring areas are imposing harmful effects on the health of people living in those areas. Hence, more clear observation of the air quality in the region is highly desirable to understand the dispersion of the enhanced pollution in the Dehradun. The major research gaps found from the study are:

- The ambient air of Dehradun city is polluted with numerous heavy metals ions and their present levels in the air has not yet been examined.
- The effect of different seasons of Dehradun city on the environmental presence of heavy metals has not yet been addressed.

- The correlation between the presence of heavy metals in the environment and their harmful effects on human health in Dehradun city has not been evaluated yet.
- Different mitigation strategies that can be employed for alleviating the heavy metal pollutants in air need discussion.

Therefore, in the present work we have exhaustively evaluated the presence of the heavy metals in the ambient air of Dehradun at different selected sites. Different season‘s data was collected for better understanding the effect of weather patterns on the environmental pollutants. The obtained results the survey study was conducted to find a relation between the presence of heavy metals in the ambient air and its harmful effect on the human health.

Finally, we have looked into different mitigation strategies that can be employed by the authorities to reduce the air pollutants in the city.

---End of Chapter 2---

CHAPTER-3

MATERIALS AND METHODS

CHAPTER 3

MATERIALS AND METHODS

Materials:

All chemicals used in the study were of analytical grade. The standard solutions of lead (Pb), cadmium (Cd), chromium (Cr), iron (Fe), mercury (Hg), copper (Cu), arsenic(As) and Selenium (Se) were purchased from Certipur® Merck Darmstadt, Germany. Air sampler AAS127 from Ecotech Instruments, India, Whatman EPM 2000 47 mm circular filter paper, Nitric acid (70 % pure) and conc. Hydrochloric acid (35-37% pure) was used. All experiments wereperformed in double distilled water.

Instruments:

Atomic Absorption Spectrophotometer (AAS) (Thermo Scientific™ iCE™ 3000 Series) was used for carrying out all the study. The Thermo Scientific™ iCE™ 3000 Series AAS as shown in figure 3.1 enables simple measurement of elements in a wide range of samples. The flame atomizer is ideal for the measurement of high concentration samples, whilst the furnace atomizer achieves sub ppb detection. The dual atomizer system with Stockdale optics incorporates advanced background correction with a unique quadline deuterium source and optional Zeeman correction to ensure accurate analysis.

Working principle of instrument: Atomic Absorption Spectrometry (AAS) is a technique for measuring quantities of chemical elements present in environmental samples by measuring the absorbed radiation by the chemical element of interest. This is done by reading the spectra produced when the sample is excited by radiation. Atomic absorption methods measure the amount of energy in the form of photons of light that are absorbed by the sample. A detector measures the wavelengths of light transmitted by the sample, and compares them to the wavelengths which originally passed through the sample. A signal processor then integrates the changes in wavelength absorbed, which appear in the readout as peaks of energy absorption at discrete wavelengths.

Figure 3. 1 Atomic Absorption Spectrophotometer (Thermo Scientific™ iCE™ 3000 Series)

The energy required for an electron to leave an atom is known as ionization energy and is specific to each chemical element. When an electron moves from one energy level to another within the atom, a photon is emitted with energy E. Atoms of an element emit a characteristic spectral line. Every atom has its own distinct pattern of wavelengths at which it will absorb energy, due to the unique configuration of electrons in its outer shell. This enables the qualitative analysis of a sample. The concentration is calculated based on the Beer-Lambert law. Absorbance is directly proportional to the concentration of the analyte absorbed for the existing set of conditions. The concentration is usually determined from a calibration curve, obtained using standards of known concentration. However, application of the Beer-Lambert law in AAS is not possible due to: variations in atomization efficiency from the sample matrix, non-uniformity of concentration and path length of analyte atoms.

Sites for sample collection:

The air samples for the study were collected from nine different localities of Dehradun city. The different locations were:

- *Location I (Sahaspur):* It‘s an industrialized area with many manufacturing units of various MNCs and other small-scale industries.
- *Location II (ISBT):* It‘s a rapidly developing area of Dehradun. The place is well connected with the roads and national highways that allow easy commuting for its residents.
- *Location III (Ballupur):* This is a residential cum market area with heavy population and witnesses a large no of vehicular movement throughout the day.
- *Location IV (Doon hospital road):* It‘s a highly crowded area having large number of vehicular movement all day.
- *Location V (Dalanwala):* This is an extensively populated area consisting of many slum areas and situated nearby the railway station.
- *Location VI (Clock Tower):* This area is the prime location and main market place of Dehradun area, consisting of many residential complexes and shopping malls and other markets drawing huge population towards it.
- *Location VII (Premnagar):* This place lies just off National Highway 72 (Chakrata Road), about 8 km from Clock Tower, Dehradun.
- *Location VIII (Jakhan):* Residential Area

- *Location IX (Selaqui Industrial area):* This area has number of industries that leads to the increment of pollutant in the city. Different locations are shown in the figure 3.2.

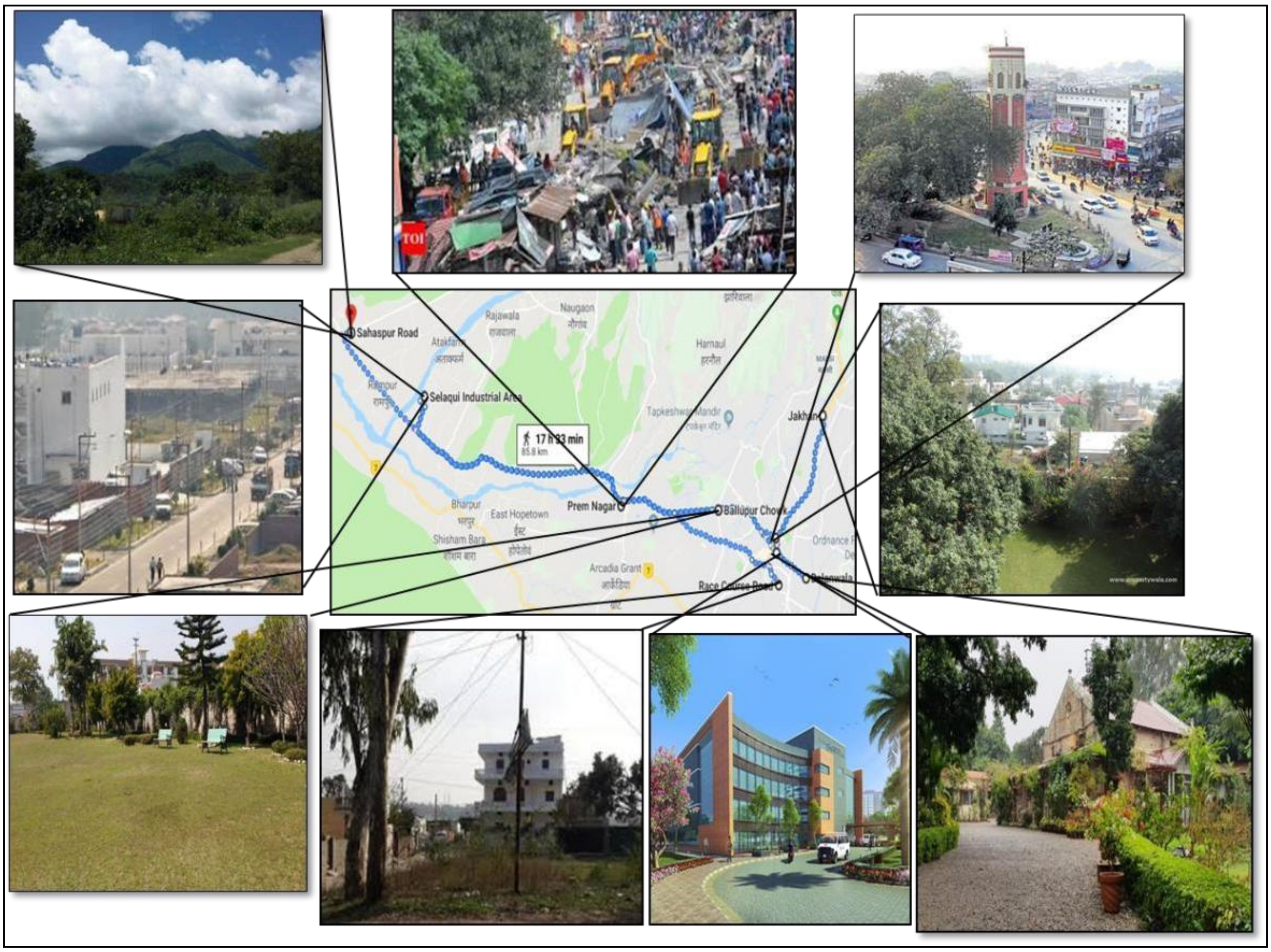

Figure 3. 2 Nine different locations selected for the study.

Quality Control:

For precision and accuracy, all the glassware‘s and filter assemblies were acid-washed and dried in hot-air oven. Only the calibrated glasswares were used. HPLC Grade de-ionized water was used for calibration of the device. Conditioning of filter paper was done by using Humidity chamber at 25 degree centigrade with 40 to 50 % of humidity.

Collection of sample:

An Ecotech PM2.5/10 air sampler (AAS127) was placed on an elevated platform of 1.5 m high at a distance of 20 m away from obstructions and used to sample the PM 10 dust from nine different locations as shown above in figure 3.2. After setting the machine, marked weighed EPM 2000 filter paper were fixed under the Air Sampler one at a time. Marked and pre-weighted filter paper was then clamped between the top cover of filter adaptor assembling of the machine. The filter cover was then closed and the machine switched on taking note of the time. The filter papers were removed after 24 hours and obtain their weight after exposure. After the measurement of the mass, the filter paper was ready for extraction to determine concentration of metal.

Figure 3.3 Air sampling unit at Premnagar site.

Method:

Sample extraction:

The extraction and analysis of heavy metals from the filter paper were conducted as per USEPA method of hot acid procedure (USEPA, 1999). To ensure the extraction of the heavy metals contained in the form of suspended particulate matters on the filter paper, the filter papers were digested with conc. Nitric acid (70 % pure) and conc.Hydrochloric acid (35-37%pure) (Melaku et al., 2008).The HNO_3 and HCl were taken 15mL and 5mL respectively and were poured in a glass beaker. The Teflon coated glass beaker was kept on the water bath for1-2 Hours at 90°C till only 3-5 mL of the acidic solution was left. The Teflon coated beaker was washed with10 mL of de-ionized water and the solution containing acid and some particulates from the filter paper was filled in a conical flask (USEPA, 2012). 20mL of de-ionized water was again added into the solution. Then the solution was filtered using Whatman 42 filter paper in a 50 mL calibrated volumetric flask and the volume was made upto the 50 mL mark of the flask with distilled water and was later stored in refrigerated condition for further testing.

Standard preparation of reference heavy metals:

The stock solutions of lead (Pb), iron (Fe), copper (Cu), cadmium (Cd), chromium (Cr), mercury (Hg), arsenic (As) and selenium (Se) were prepared using Certipur® standard solution (1000 ppm). The stock solutions were further used to make standard solutions of 0.50 ppm, 1.00 ppm, 2.00 ppm, 3.00 ppm and 10.00 ppm by serial dilution methods from the stock solution. The standard solutions thus prepared were shaken and the flasks were made upto 100 mL marks.

Sample analysis:

The digested sample solutions were analyzed for the heavy metals (Pb, Cu, Fe, Cd, Cr, Hg, As and Se) using the Atomic Absorption Spectrophotometer (Thermo Scientific iCE 3000 Series). The calibration plot method was used for the analysis. The air-acetylene gas was used for the analysis of Pb, Cu, Fe, Cd, Hg, As and Se while nitrous-oxide gas along with air-acetylene gas was used for the analysis of Cr. The wavelengths for the determination of the elements were Cu 324.8, Cd 228.8, 248.3 Fe 248.3, Hg 253.7, Pb 283.3, As 193.7, Se

196.0 and Cr 357.9 respectively. The concentrations of heavy metals in the solutions were displayed in ppm by the AAS. The concentration of the heavy metals was analyzed using AAS (Thermo ScientificiCE 3000 Series). For quality control, standard stock solutions were prepared for each metal. The standard Reference Materials (SRMS) were prepared using Certipur® standard solution (1000ppm).

NOTE: This research work was done at UPES HSE NABL Accredited Laboratory (Certificate no. TC-5420) so all the equipment was calibrated as per NABL Guidelines.

---End of Chapter 3---

CHAPTER-4

RESULTS AND DISCUSSION

CHAPTER 4

RESULTS AND DISCUSSION

Prevalence of heavy metals in the ambient air of Dehradun:

The heavy metal concentration in the particulates was found by digesting the filter paper and analyzing the presence of the metals in the samples using the atomic absorption spectrophotometer. Table 4.1-4.3 provides the concentration of Pb, Cu, Fe, Cd, Cr, Hg, As and Se at nine different locations of Dehradun and the data at these locations was collected since February 2017 to January 2018. The concentration of all heavy metals taken into consideration had been considered toxic, if the concentration in air are found beyond permissible safe limits set by different environmental agencies.

Concentration of lead (Pb):

Lead is an important naturally occurring bluish-grey metal, found in a low concentration in the earth crust. It is insoluble in water but, its compounds are soluble. The main use of lead is in making batteries. Lead is applied hugely in the production of metal products, including sheet lead, solder, and pipes, and in ceramic glazes, cable covering, and other products. Tetraethyl lead was also used in gasoline earlier to increase the octane rating until lead additives were phased out and they got banned from use in gasoline in the U.S. by the EPA in 1996.

Ambient Air Concentrations: Urban concentrations of lead average 1,100 ng/m^3, non-urban concentrations average 210 ng/m^3, and remote areas average 20 ng/m^3 (HSDB 2010). The standard limit defined by the CPCB Air (ambient) 1.0 µg/m^3.

Table 4. 1 Concentration of heavy metals in ambient air of Dehradun from February – May 2017

Sr. No	Dehradun Location	Pb ($\mu g/m^3$)	Cu ($\mu g/m^3$)	Fe (mg/m^3)	Cd ($\mu g/m^3$)	Hg ($\mu g/m^3$)	Cr (ng/m^3)	As (ng/m^3)	Se (ng/m^3)
1.	Sahaspur	1.98	0.06	0.03	0.21	0.14	0.05	21.0	0.60
2.	ISBT	0.49	0.03	0.01	0.06	0.11	0.04	16.60	7.71
3.	Ballupur	0.49	0.05	0.05	0.07	0.14	0.04	18.85	7.05
4.	Doon hospital road	0.47	0.07	0.05	0.07	0.08	0.04	24.06	10.39
5.	Dallanwala	0.89	0.07	0.0761	0.122	0.142	0.052	15.28	14.62
6.	Ghantaghar	1.0381	0.2515	0.2067	0.382	0.141	0.046	27.42	17.99
7.	Premnagar	0.56	0.2565	0.1905	0.373	0.141	0.050	27.55	17.82
8.	Jakhan	0.2542	0.078	0.008	0.066	0.072	0.044	14.89	0.46
9.	Selaqui Industrial area	2.6094	0.1346	0.0396	0.327	0.144	0.061	21.89	0.979

Table 4. 2 Concentration of heavy metals in ambient air of Dehradun from June– September 2017

Sr. No	Dehradun location	Pb ($\mu g/m^3$)	Cu ($\mu g/m^3$)	Fe (mg/m^3)	Cd ($\mu g/m^3$)	Hg ($\mu g/m^3$)	Cr ($\mu g/m^3$)	As (ng/m^3)	Se ($\mu g/m^3$)
1.	Sahaspur	1.38	0.06	0.03	0.14	0.14	0.05	15.75	0.14
2.	ISBT	0.25	0.03	0.01	0.06	0.11	0.04	16.05	7.24
3.	Ballupur	0.25	0.04	0.05	0.06	0.14	0.04	16.51	6.50
4.	Doon hospital Road	0.47	0.05	0.05	0.05	0.08	0.04	23.34	9.75
5.	Dallanwala	0.81	0.05	0.05	0.07	0.14	0.04	14.37	13.98
6.	Ghantaghar	0.51	0.22	0.13	0.29	0.14	0.04	26.32	16.85
7.	Premnagar	0.79	0.18	0.16	0.27	0.13	0.04	25.79	16.82
8.	Jakhan	0.15	0.07	0.06	0.04	0.06	0.03	13.62	0.42
9.	Selaqui Industrial area	2.20	0.11	0.03	0.19	0.14	0.05	18.91	0.15

Table 4. 3 Concentration of heavy metals in ambient air of Dehradun from October 2017- January 2018

Sr. No	Dehradun location	Pb ($\mu g/m^3$)	Cu ($\mu g/m^3$)	Fe (mg/m^3)	Cd ($\mu g/m^3$)	Hg ($\mu g/m^3$)	Cr (ng/m^3)	As (ng/m^3)	Se (ng/m^3)
1.	Sahaspur	2.37	0.06	0.03	0.39	0.13	0.05	21.07	0.31
2.	ISBT	0.40	0.04	0.01	0.07	0.11	0.05	17.66	8.40
3.	Ballupur	0.49	0.03	0.05	0.10	0.14	0.05	19.14	7.15
4.	Doon hospital road	0.51	0.08	0.05	0.06	0.08	0.05	24.07	10.43
5.	Dallanwala	0.89	0.07	0.07	0.13	0.14	0.05	15.52	14.83
6.	Ghantaghar	1.73	0.25	0.27	0.44	0.14	0.04	28.09	18.54
7.	Premnagar	1.18	0.35	0.20	0.37	0.14	0.05	27.57	17.97
8.	Jakhan	0.35	0.08	0.00	0.08	0.076	0.04	14.94	0.25
9.	Selaqui Industrial area	2.67	0.23	0.04	0.44	0.14	0.06	22.01	0.22

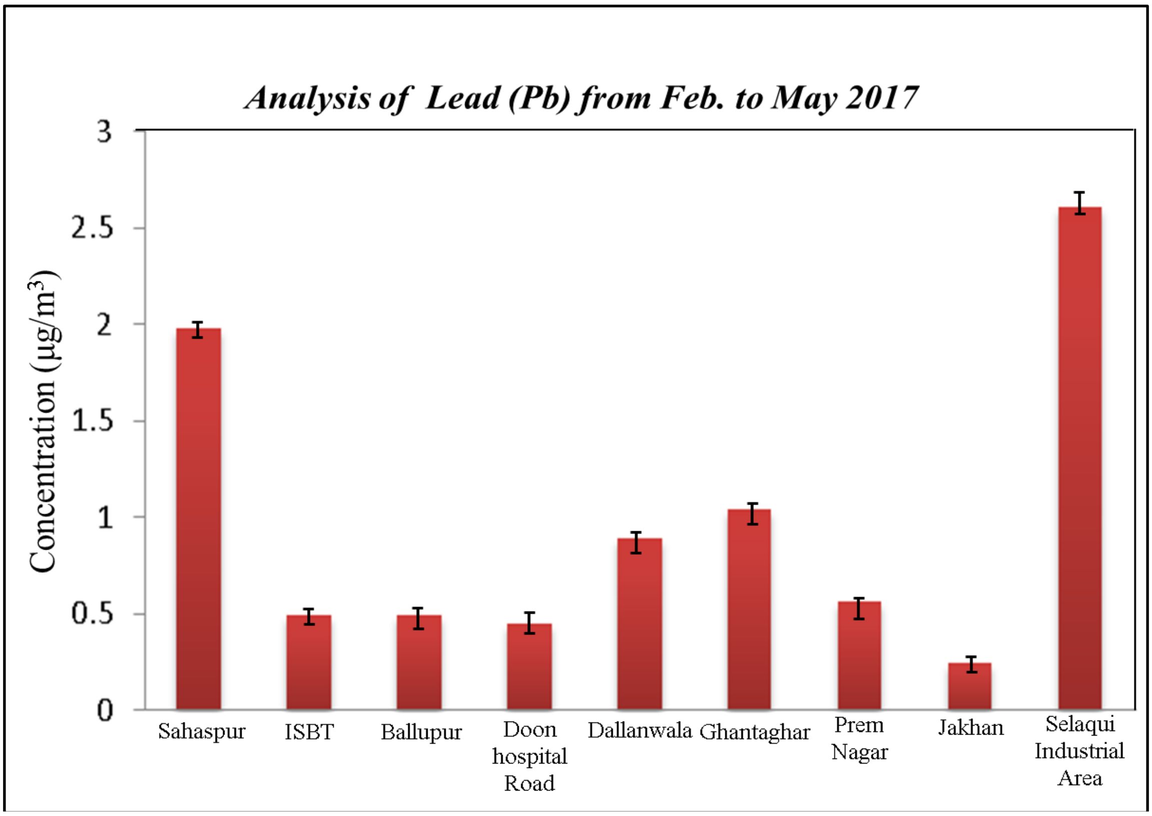

Figure 4. 1 Concentration of lead at different locations from Feb. to May 2017

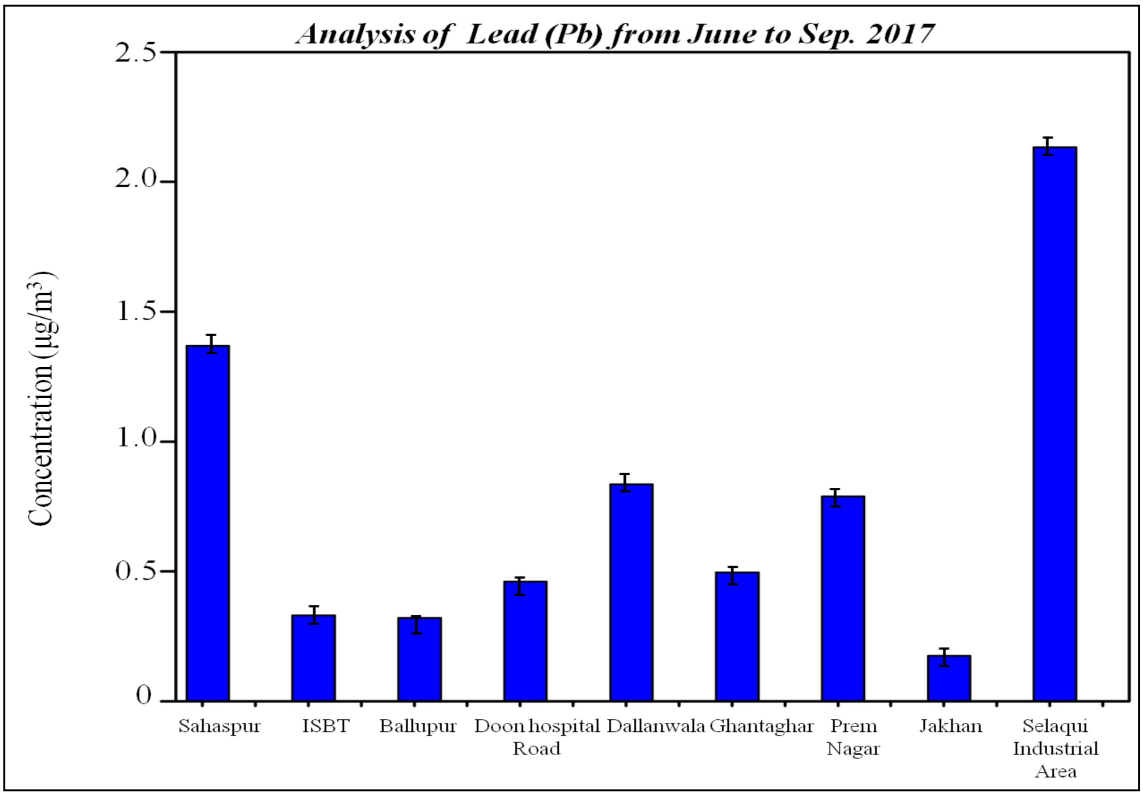

Figure 4. 2 Concentration of lead at different locations from June to Sep. 2017

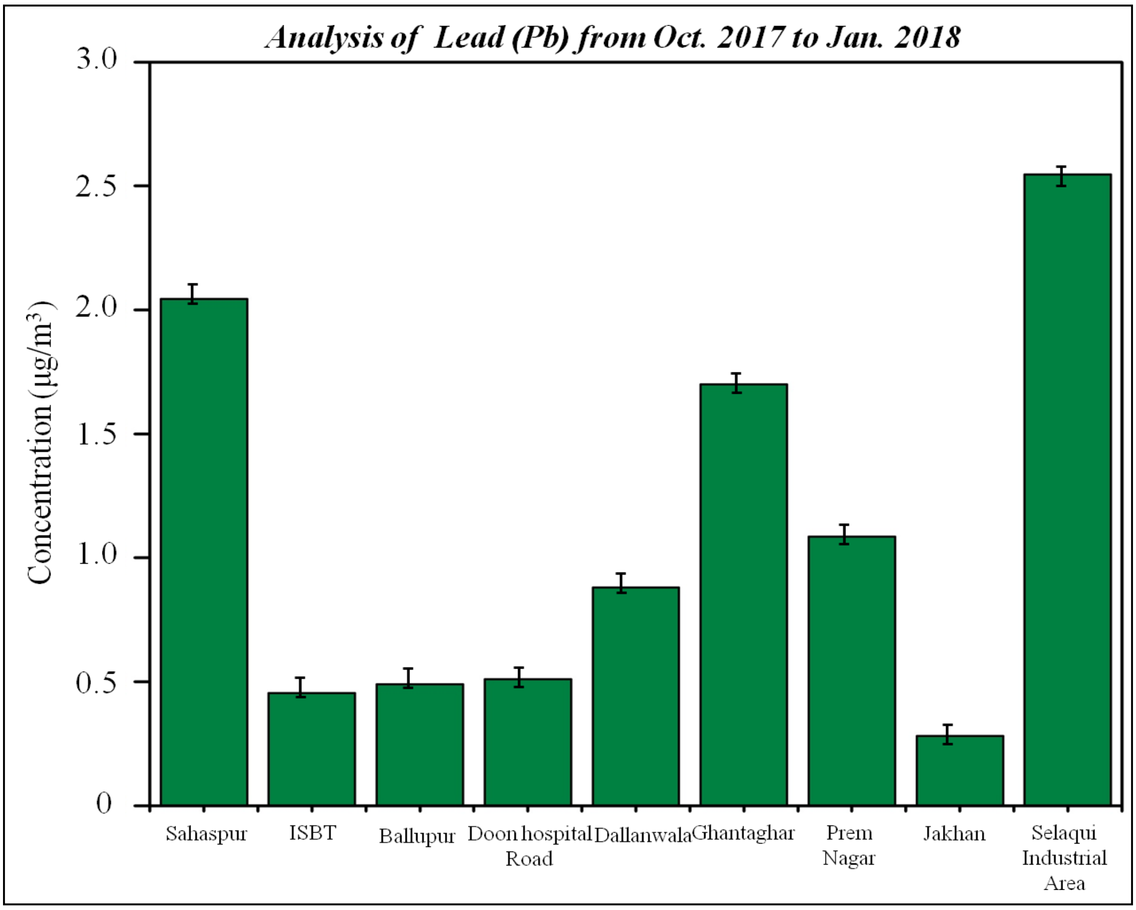

Figure 4. 3 Concentration of lead at different locations from Oct. 2017 to Jan. 2018

Concentrations of Pb at the different locations in the ambient air have been shown in the bar graphs (figure 4.1-4.3). The data obtained from the study lead to a confirmation that among different nine locations of study, the concentration of Pb was highest at the Selaqui Industrial Area (2.60 μgm^{-3}) during the period of study from February to May 2017 followed by Sahaspur, Dallanwala and Prem Nagar as depicted in the graphical figure 4.1. Furthermore, it was observed that the concentration was also found to be elevated in two other different seasons i.e. June to September 2017 and October 2017 to January 2018 as shown in figure 4.2 and 4.3, respectively. The concentration of lead obtained in the study was much higher in all the seasons than the set guidelines of various environmental agencies, hence, these areas have been considered polluted with high concentration of Pb which might lead to different health problems for the population residing in those areas.

Short-term health effects: Lead is highly toxic even in low doses in blood, lead in children may cause death. Short-term exposure includes problems like brain damage, kidney damage, and gastrointestinal damage.

Long-term health effects: Long-term effects of lead can bring the blood, central nervous system, blood pressure, kidneys, and Vitamin-D metabolism. Neurological symptoms have been reported in the workers in the field. In adults, it may slow down the nerve conduction process. Furthermore, its exposure can also have effect on the reproductive system, for example, miscarriages, decreased sperm count, low birth weight, slowed postnatal neurobehavioral development (EPA, 2017).

Cancer risk: EPA categorized it to be a Group B2, human carcinogen.

Concentration of Copper (Cu):

Copper usage is mainly for making wire, plumbing pipes, and sheet metal. Copper is also mixed with other metals in making brass and bronze pipes. Its compounds are also employed in agriculture to treat plant diseases such as mildew, for the treatment of water and, also as preservatives for wood, leather, and fabrics.

Ambient air concentration: Cu concentration was measured in the ambient air at different location of Dehradun as illustrated in the figures 4.4-4.6. The obtained data confirmed that among different nine locations of study, the concentration of Cu was higher in the Ghantaghar area and Prem Nagar area measuring 0.251 and 0.256 μgm^{-3}, respectively from February to May 2017. Similarly, it was found that the concentration was also higher in the same localities during June to September 2017 and October 2017 to January 2018 as shown in figure 4.5 and 4.6, respectively. The standard limit defined by the USEPA for the Cu is [Air (ambient) 2 μgm^{-3}].

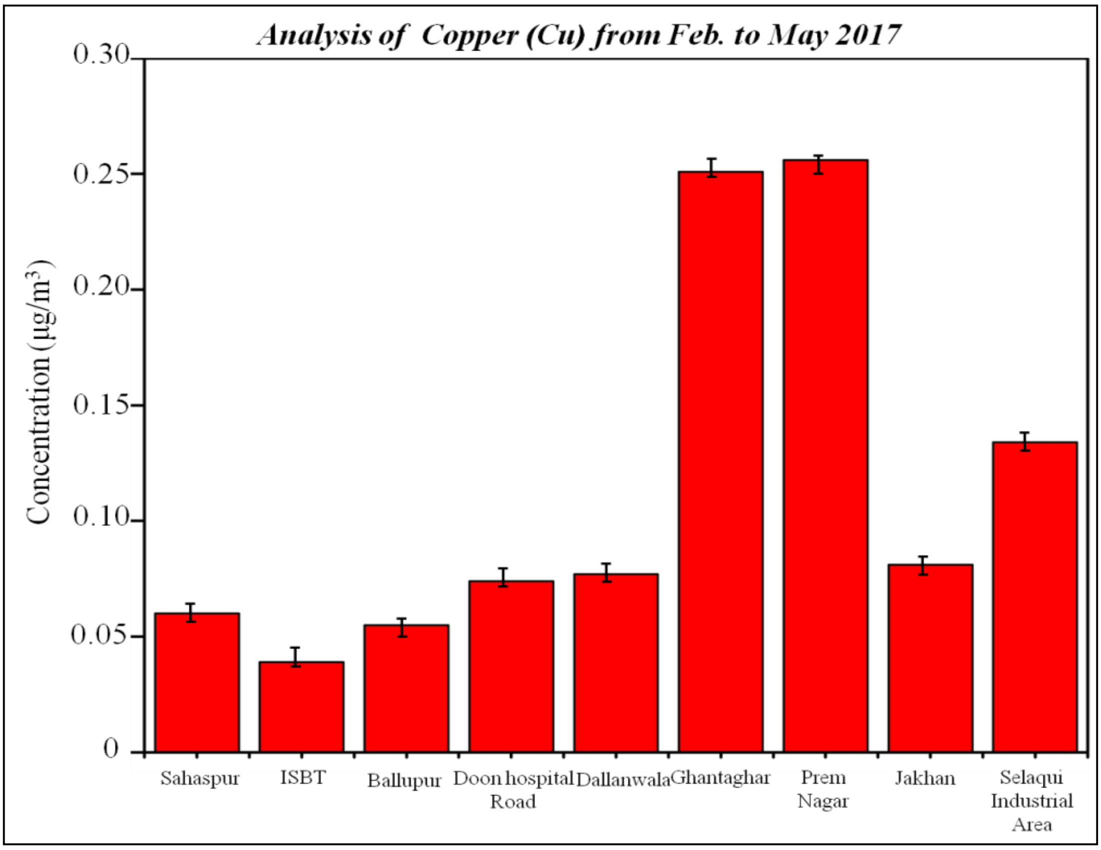

Figure 4. 4 Concentration of copper at different locations from Feb. to May 2017.

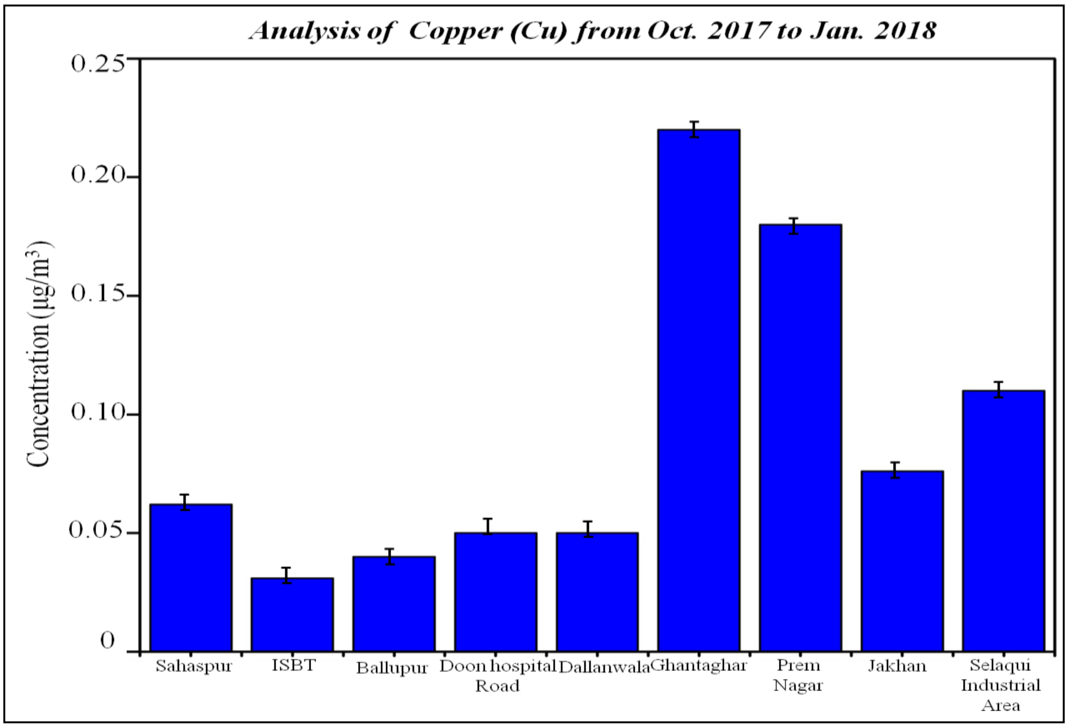

Figure 4. 5 Concentration of copper at different locations from June to Sep. 2017.

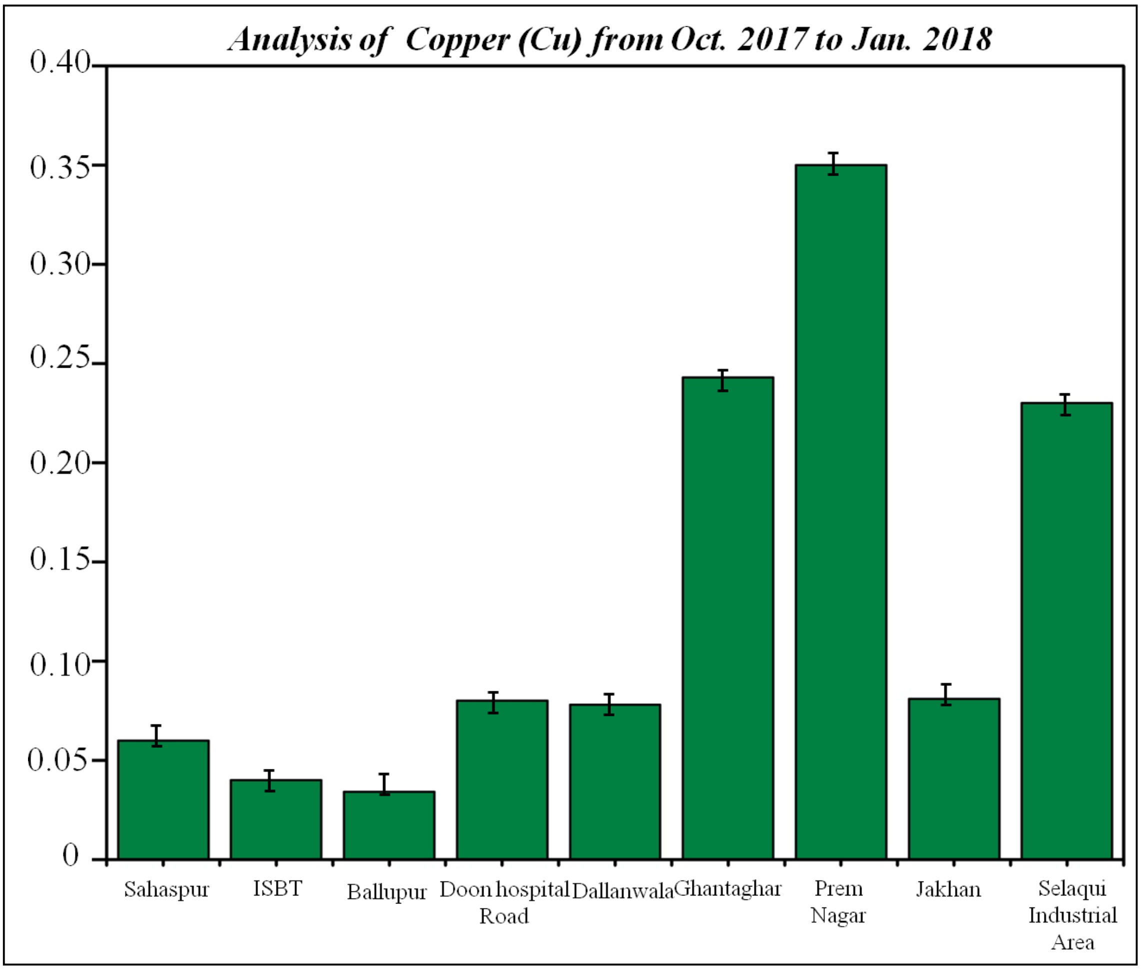

Figure 4. 6 Concentration of copper at different locations from Oct. 2017 to Jan. 2018.

Short-term Health Effects: The inhalation of dusts of copper salts can cause the irritation of nasal mucous membranes, eyes, leading to nausea, and fever. Acute copper poisoning may lead to liver injury, methemoglobinemia, and hemolytic anemia.

Long-term Health Effects: In humans, chronic overexposure to this metal ion can cause damage to the liver and kidneys and may lead to anemia (ASTDR, 2004).

Cancer Risk: Copper is currently in category Group D, not classifiable as to carcinogenicity in humans.

Concentration of Iron (Fe):

Iron is considered as an essential element required by all forms of life on earth. In addition, it occurs naturally in all foods of plant or animal origin, and is also present in food as iron oxides, as organic complexes, such as in hemoglobin.

Ambient Air Concentrations: An average iron concentration in urban air 1.6 μg/m^3 have been reported, other sources provide a range of atmospheric iron concentration of 0.9 to 1.2 μg/m^3, as particular ferric oxide. The concentration of Fe at all nine different locations was studied and the data obtained provide inference that among all the locations of study, the concentration of Fe was higher at the Ghantaghar site and Prem Nagar area during whole period as shown in figure 4.7-4.9. The concentration of iron was found much higher in these regions during all the seasons than the standard limit defined by the USEPA (1.6 mg/m^3) guidelines and hence, the areas having high concentration of Fe may cause some serious health conditions to people residing there. Therefore, it is suggested that some mitigation strategies should be applied in these areas to bring the concentration of iron as discussed below in the mitigation methods.

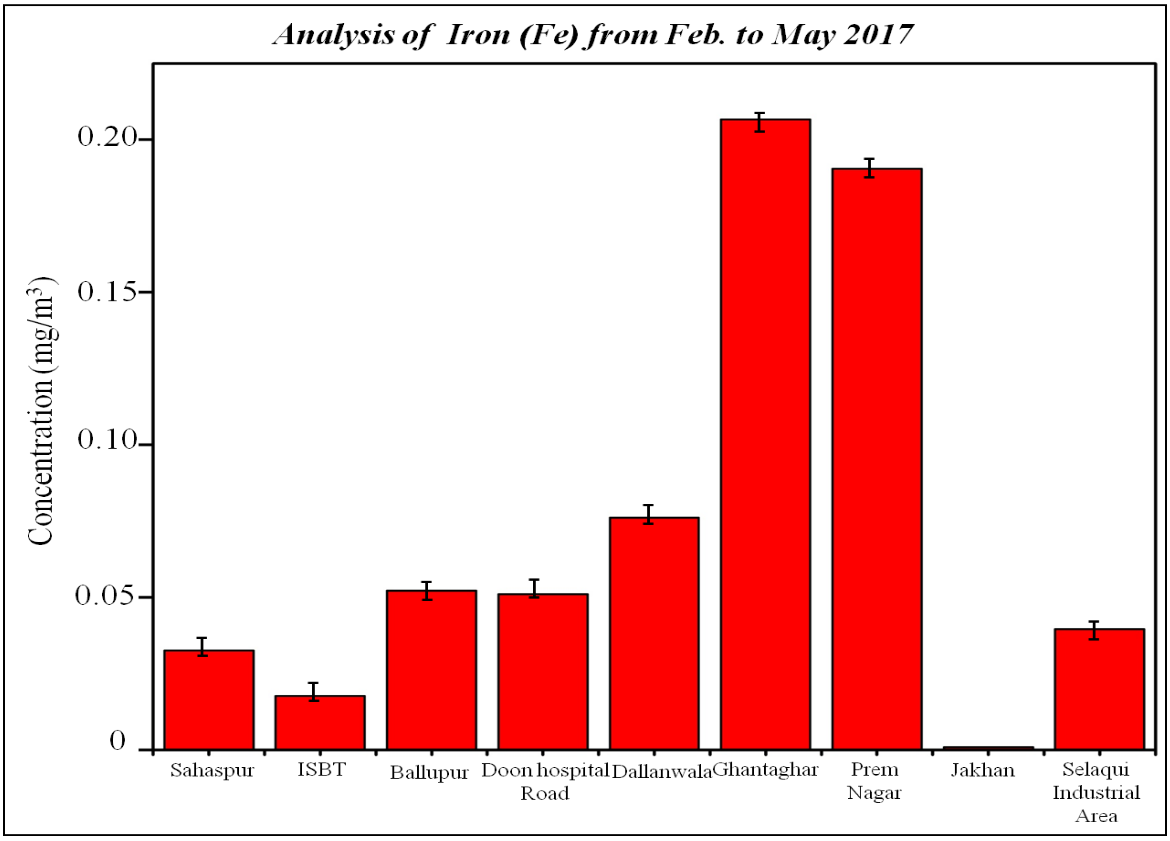

Figure 4. 7 Concentration of iron at different locations from Feb. to May 2017.

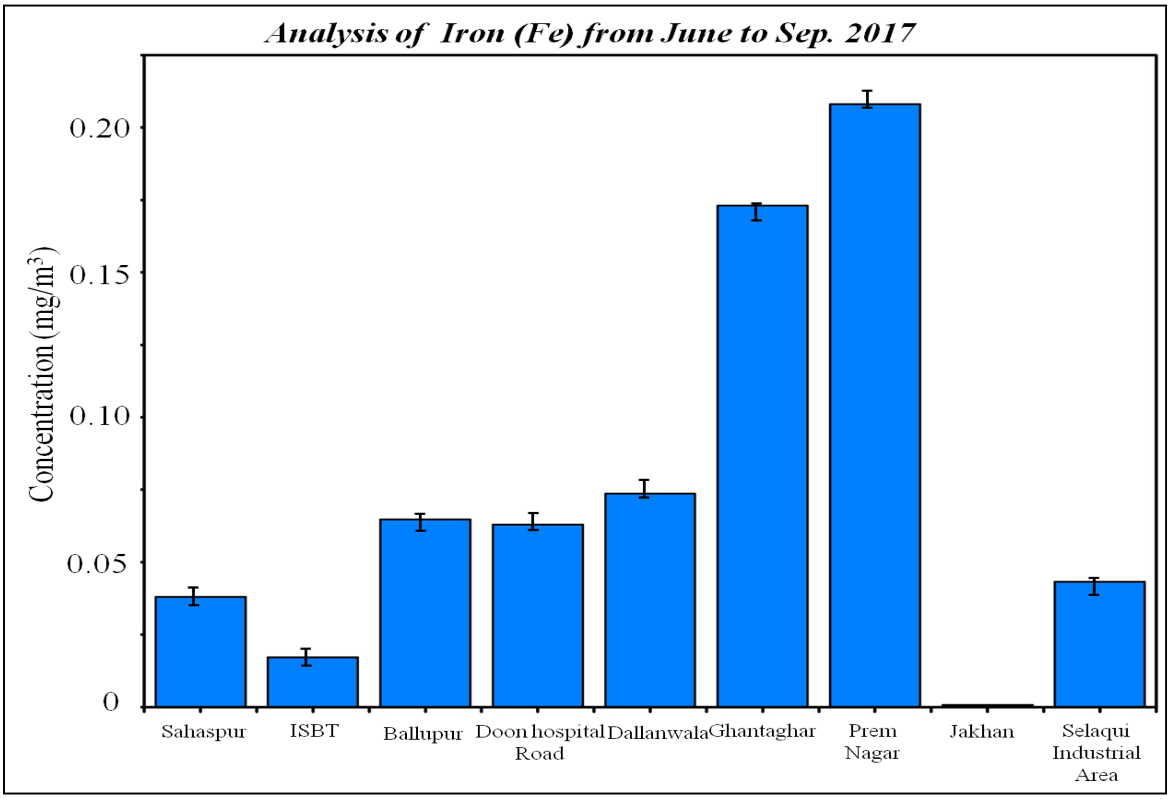

Figure 4. 8 Concentration of iron at different locations from June to Sep. 2017.

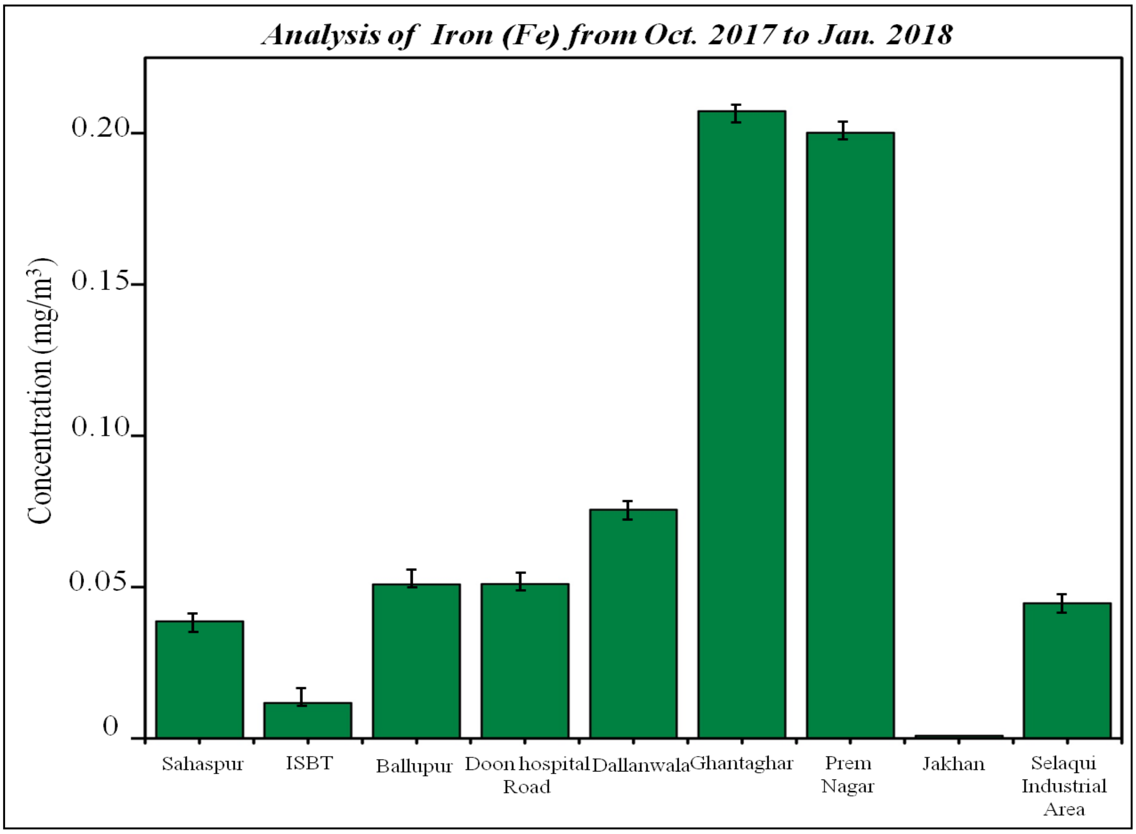

Figure 4. 9 Concentration of iron at different locations from Oct. 2017 to Jan. 2018.

Short-term health effects: Toxicity caused due to the acute iron overdose results in corrosive effects on the gastrointestinal mucosa. Inhalation of ferric salts as dusts & mists leads to irritation of the respiratory tract. Moreover, salts of ferric act as skin irritants. Major symptoms of acute iron toxicity are diarrhea, fever, nausea, stomach pain or cramping, vomiting. The other major symptoms that may develop include bluish-colored lips, fingernails, drowsiness, pale skin, seizures, and tiredness.

Long-term health effects: The corrosive effects of iron may cause stomach and intestinal erosions and ulceration. Though, there is an insufficient correlation between the severity of intestinal damage and death.

Cancer Risk: Free iron is a pro-oxidant that may induce oxidative stress and damage DNA.

Concentration of cadmium (Cd):

Cadmium is a silver-white metal found on the Earth. It is most commonly a byproduct resulting from the smelting of zinc, lead, or copper ores. Its mainly used in making of the pigments and batteries, metal-plating, and also in some plastics industries.

Ambient air concentrations: Ambient air concentration of cadmium have generally been estimated to range from 5 $\mu g/m^3$ in rural areas, from 2 to 15 $\mu g/m^3$ in urban areas, and from 15 to 150 $\mu g/m^3$ in industrialized areas (ICdA 2009). The concentration of the cadmium was found significantly low at all the sites of study in Dehradun as depicted in the figure 4.10-4.12.

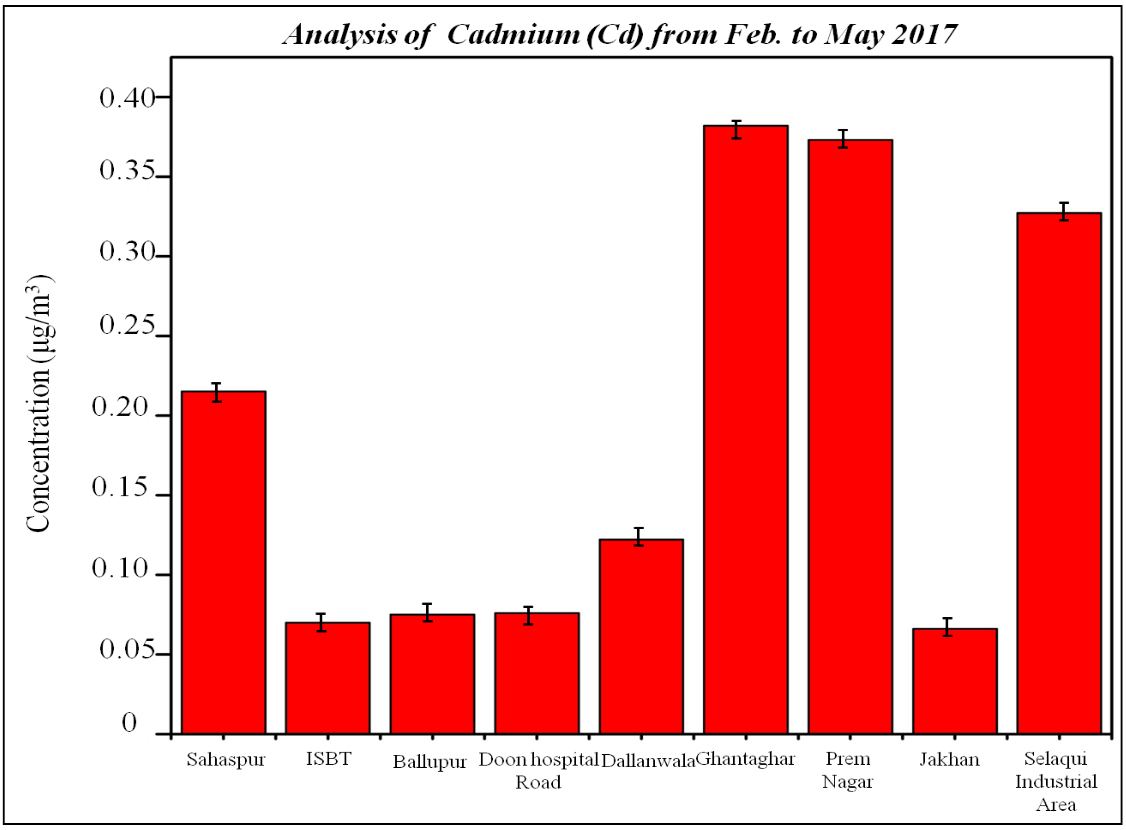

Figure 4. 10 Concentration of cadmium at different locations from Feb. to May 2017.

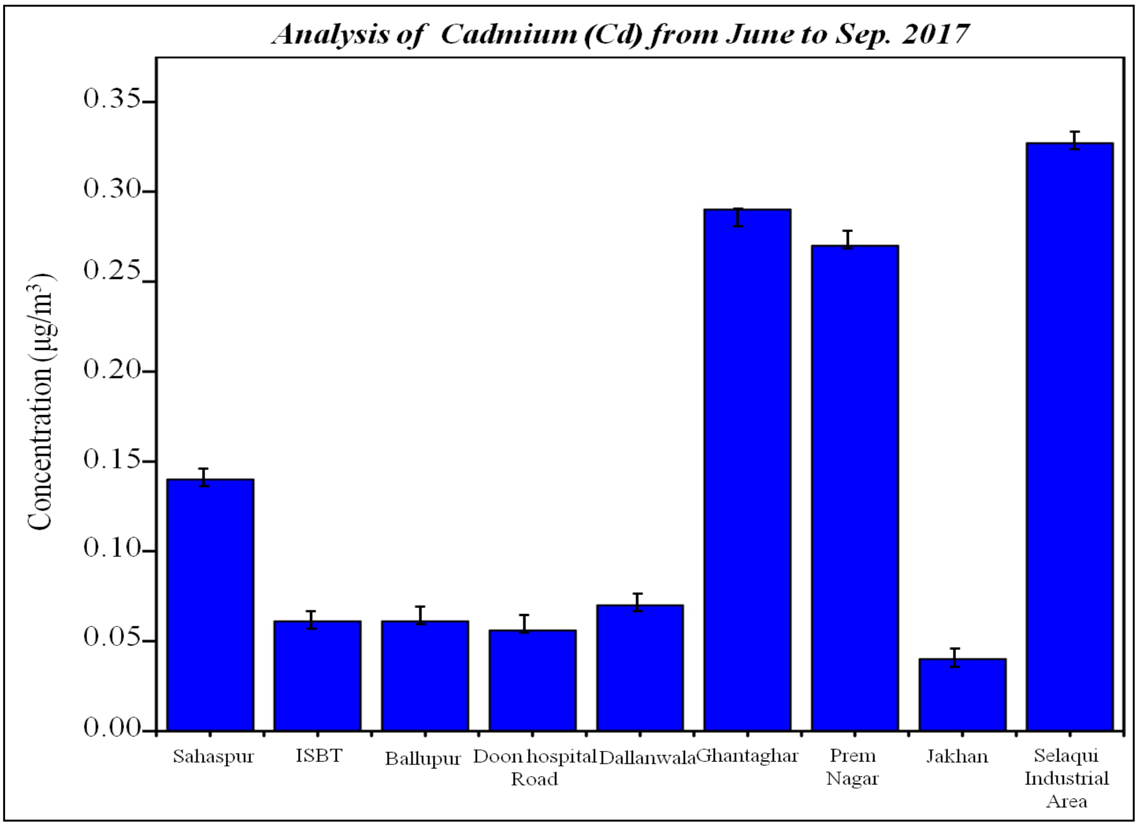

Figure 4. 11 Concentration of cadmium at different locations from June to Sep. 2017.

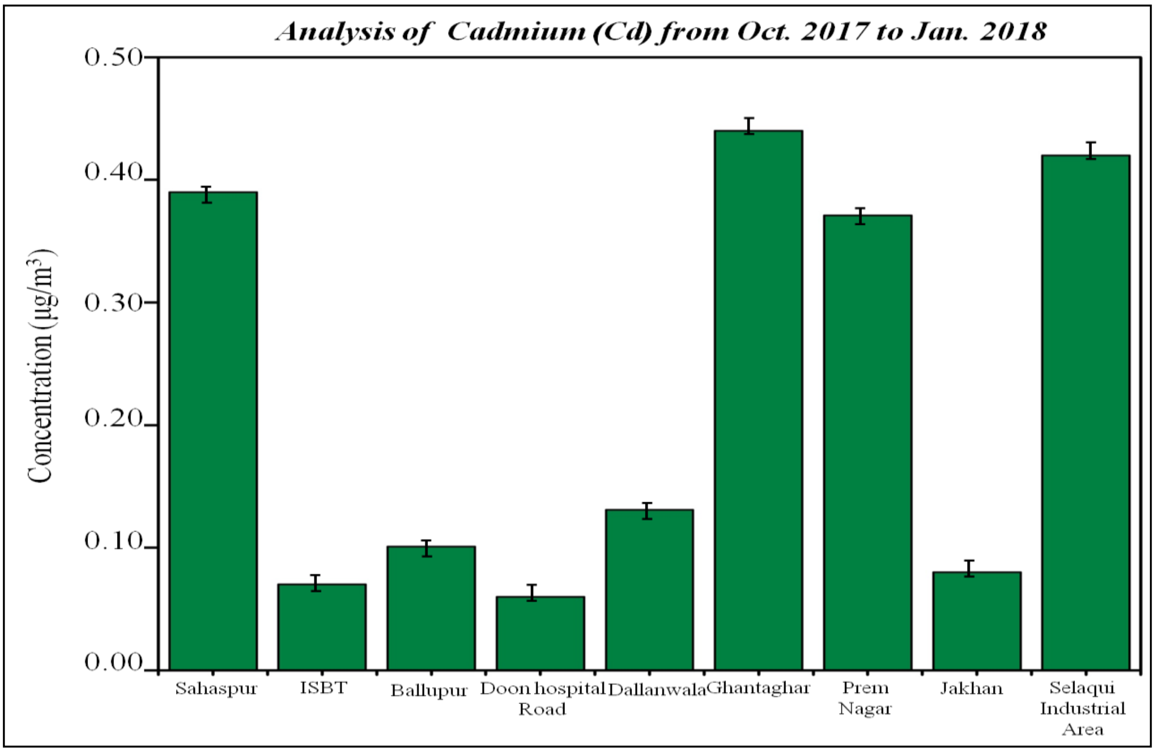

Figure 4. 12 Concentration of cadmium at different locations from Oct. 2017 to Jan. 2018.

Short-term health effects: The short-term effects include effects on the lungs like bronchial and pulmonary irritation while an acute exposure of cadmium can cause long-lasting impairment of lungs.

Long-term health effects: Long-term effects of cadmium on human can be due to inhalation and ingestion which can lead to build-up of cadmium in the kidneys, liver, lung, bone, immune system, blood, and nervous system. Some animal studies have shown fetal malformations and other developmental problems as a result of cadmium exposure.

Cancer Risk: Cadmium is a Group B1, human carcinogen as categorised by the EPA.

Concentration of mercury (Hg):

All the three forms of mercury are highly toxic to the human whether it is elemental mercury, inorganic mercury, and organic mercury. Elemental mercury finds application in the thermometers, barometers and other pressure-sensing devices.

Ambient air concentration: The reference concentrations recommended by the USEPA amounts to 0.3 µg/m^3 (IRIS, 1995). In the study, the concentration of mercury was found below permissible limits in all the regions of data collection as shown in figure 4.13-4.15 which proves that areas of Dehradun are not polluted by this element.

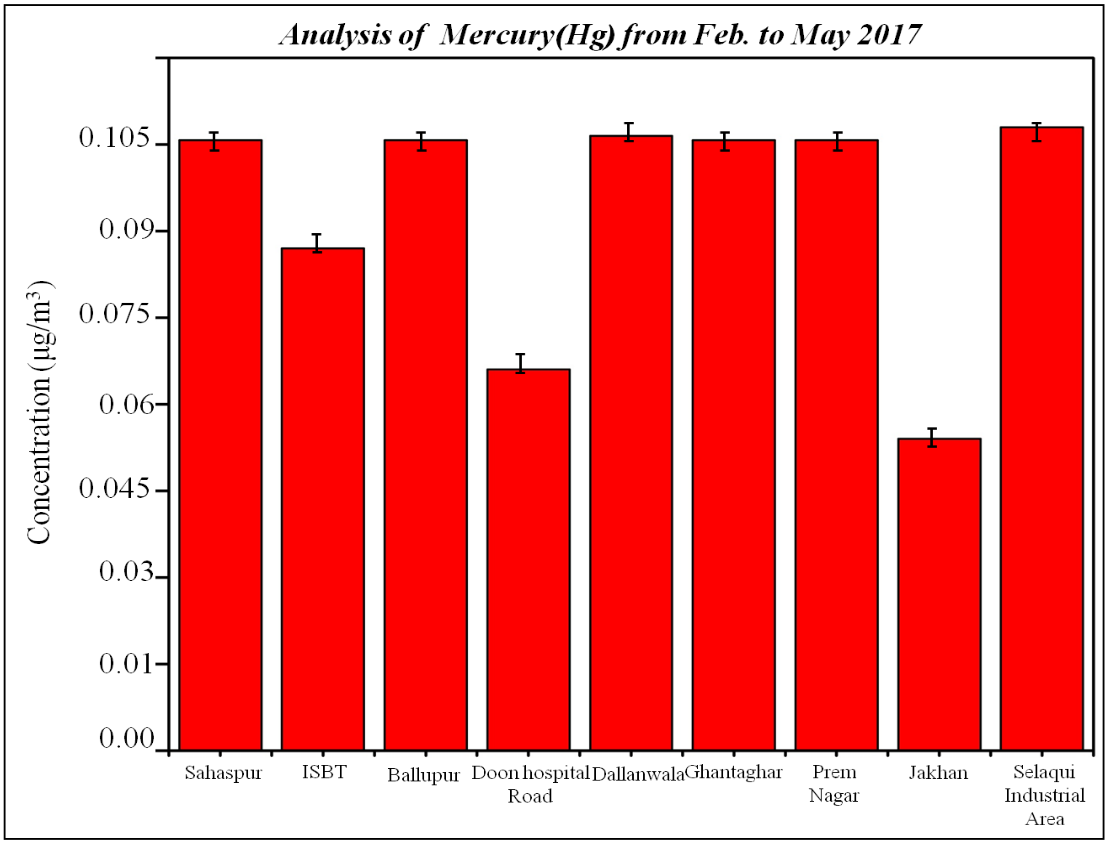

Figure 4. 13 Concentration of mercury at different locations from Feb. to May 2017.

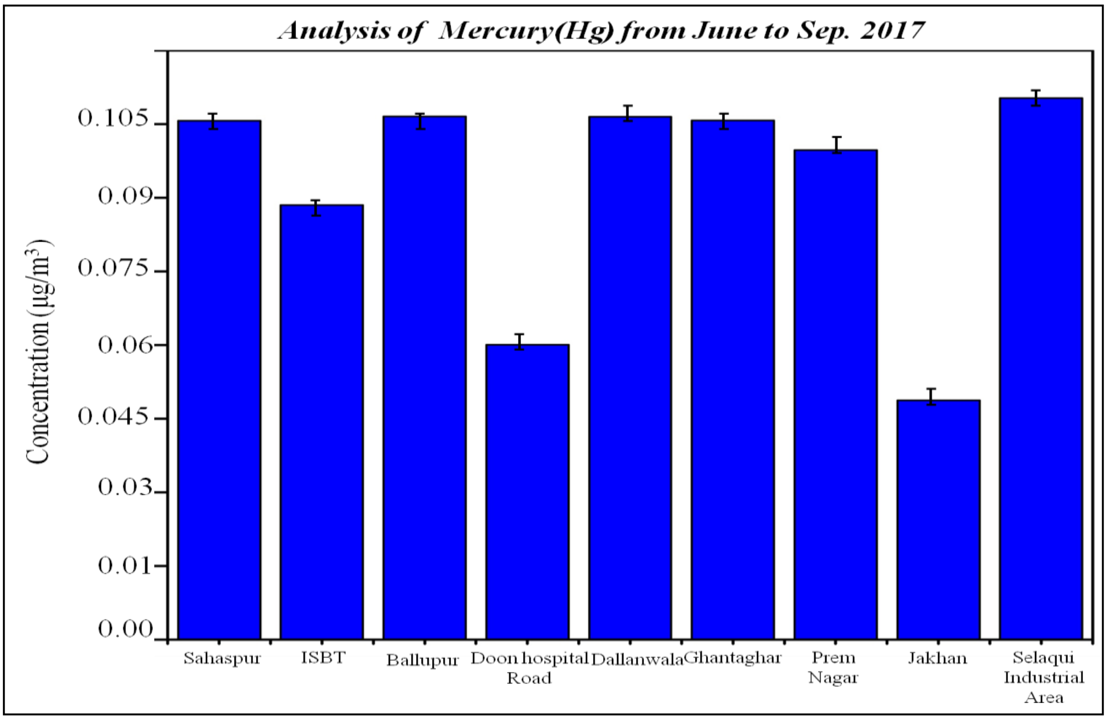

Figure 4. 14 Concentration of Mercury at different sites from June to Sep. 2017.

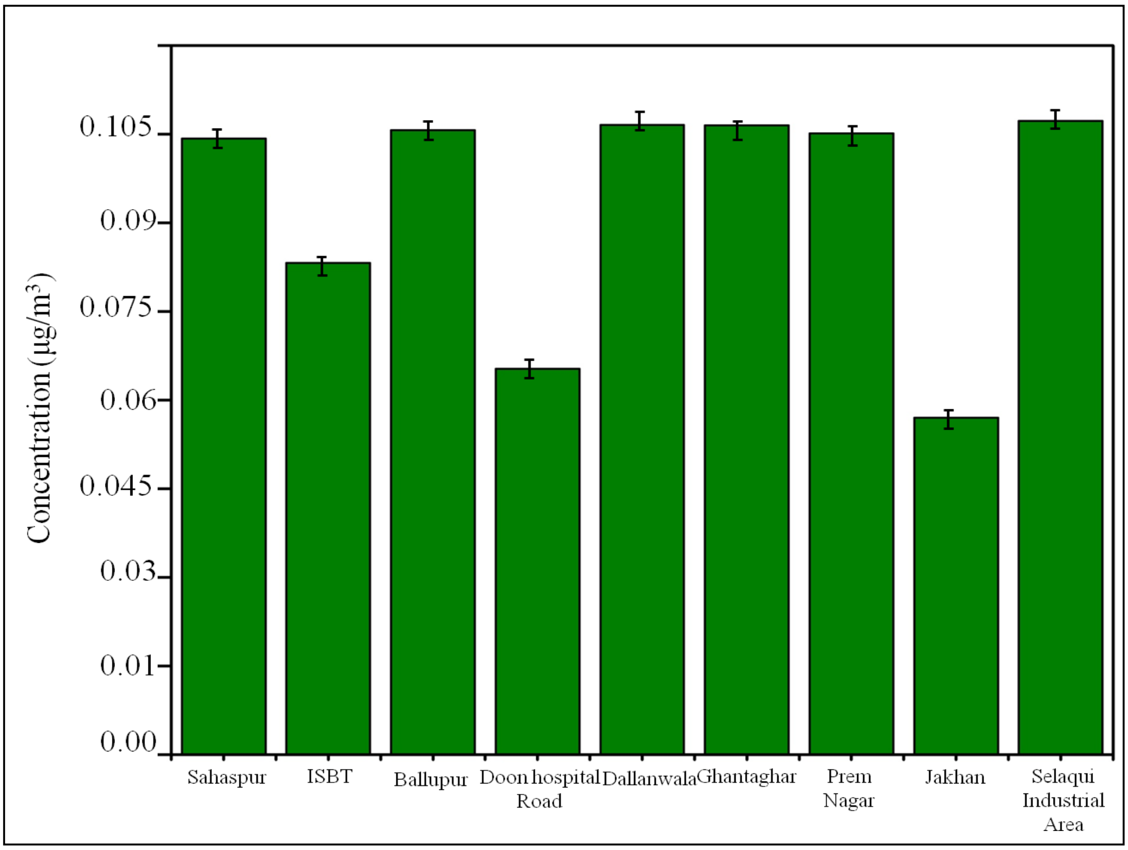

Figure 4. 15 Concentration of mercury at different locations from Oct. 2017 to Jan. 2018.

Short-term health effects: Short term exposure of high levels of elemental mercury can cause damage to CNS that may lead to tremors, mood changes, and slowed sensory and motor nerve function. Likewise, inorganic mercury exposure can also cause nausea, abdominal pain and vomiting.

Long-term health effects: Long-term exposure to elemental mercury damages CNS causing irritability, tremors and excessive shyness. Long-term exposure of the inorganic mercury can lead to kidney damage, high resorption rates, and abnormalities in fetal development. Methyl mercury, on the other hand, can cause CNS effects with symptoms like paresthesia, blurred vision, difficulty in speaking, and constriction of the visual field (EPA, 2020).

Cancer Risk: Elemental mercury is a Group D, not a human carcinogen while inorganic and methyl mercury has been classified as a Group C, possible human carcinogen.

Concentration of chromium (Cr):

Chromium is a steel-gray solid metal that has high melting point. It is also used in the steel manufacturing as well as its alloys, and its compounds both in the form of chromium (III) or chromium (VI) are utilized for chrome plating, dyes manufacturing, leather and wood preservation, and water treatments.

Ambient air concentration: The maximum chromium level measured in ambient air near a chromate manufacturing plant in Corpus, Christi, Texas was 5,500 ng/m^3 with an annual average concentration in ambient air of 400 ng/m^3. The permissible limit of chromium VI at 8-hour time-weighted average exposure is 5 µg/m^3 as per OSHA (ASTDR, 2013). The concentration of chromium was found significantly low at all sites of study in Dehradun as illustrated in the figure 4.16-4.18. The study confirms that Dehradun city is not polluted by chromium metal.

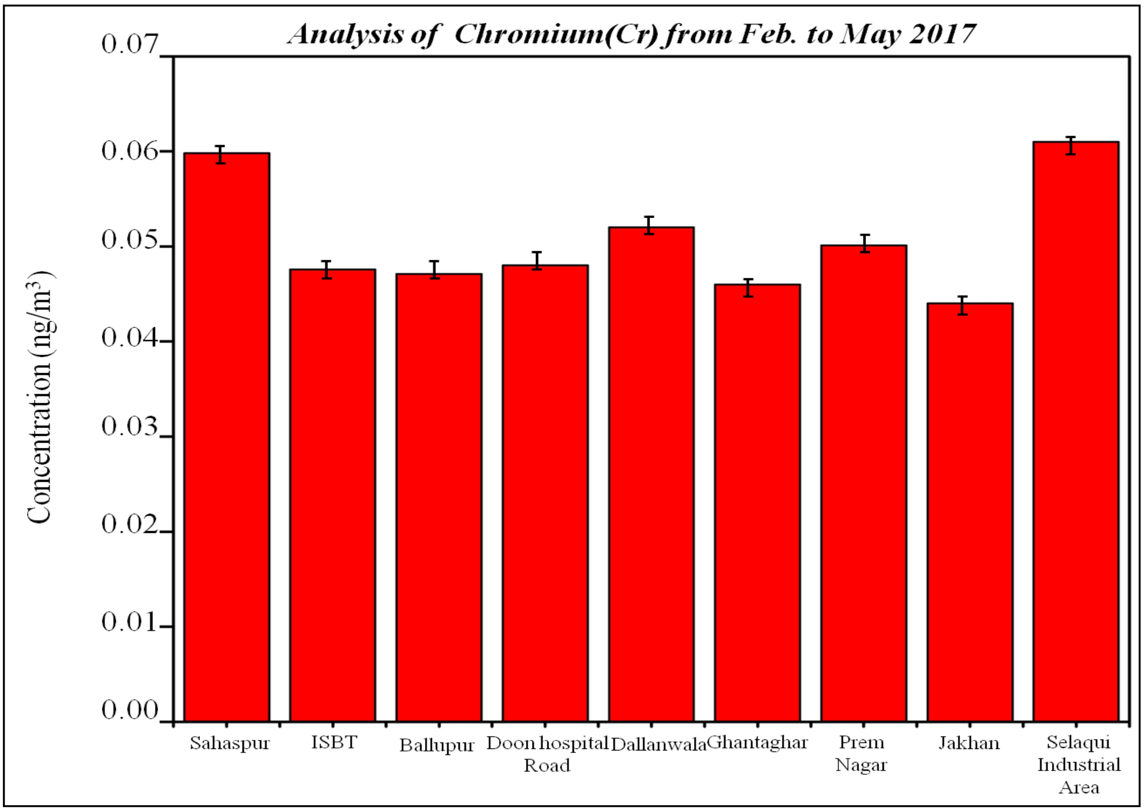

Figure 4. 16 Concentration of chromium at different locations from Feb. to May 2017.

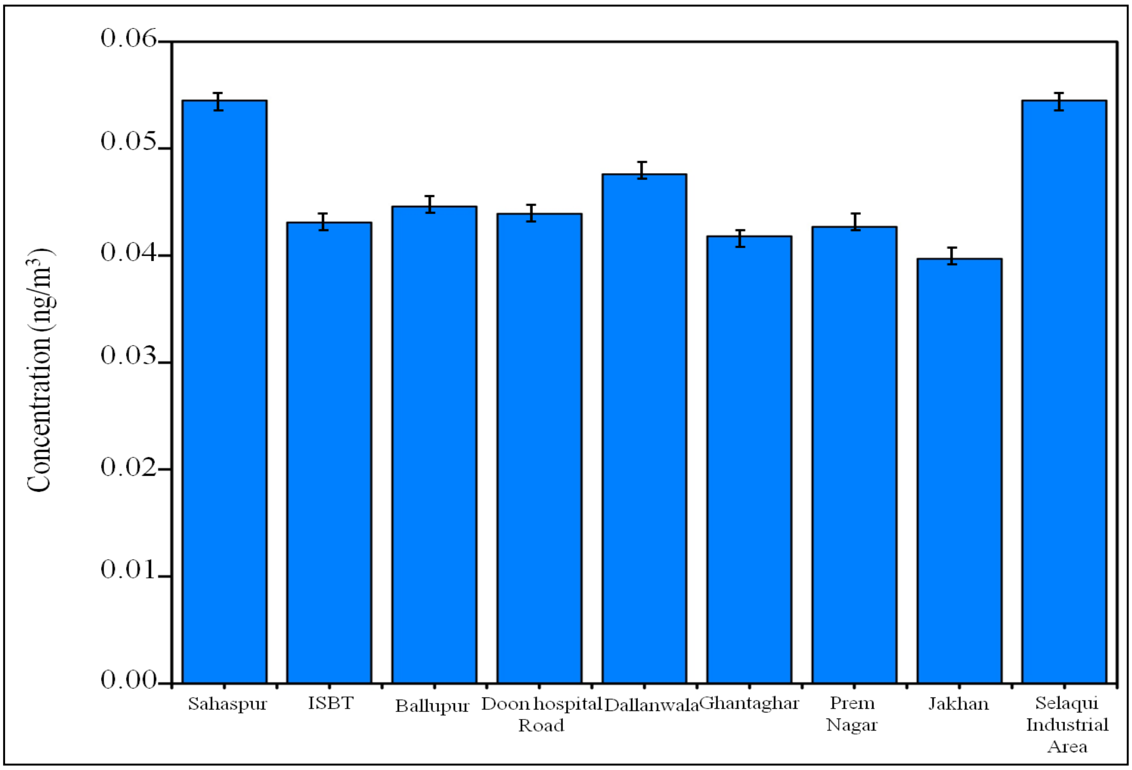

Figure 4. 17 Concentration of Chromium at different locations from June to Sep. 2017.

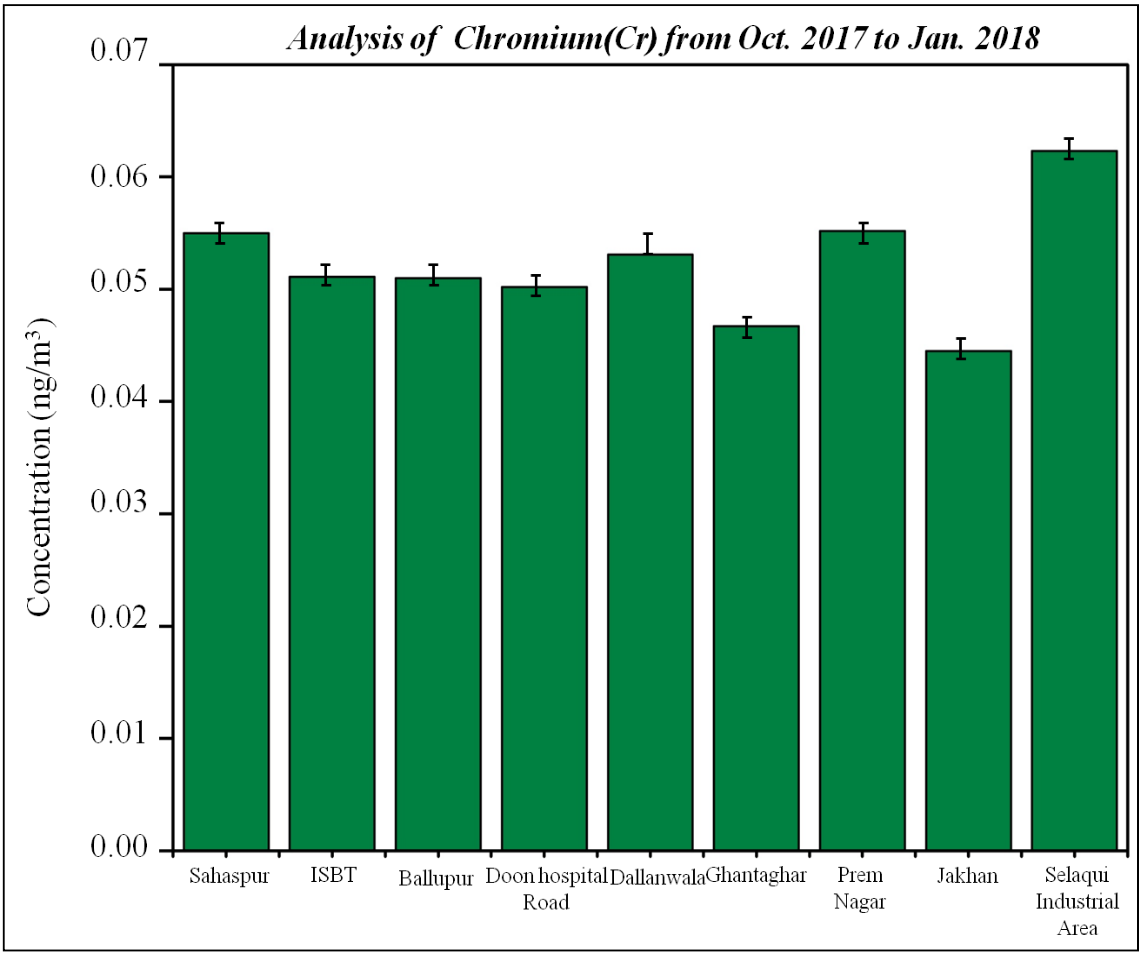

Figure 4. 18 Concentration of chromium at different locations from Oct. 2017 to Jan. 2018.

Short-term health effects: The short-term effects of chromium (VI) inhalation mainly include breath problems, and many other effects on the respiratory tract (Olaguibel and Basomba, 1989). Chromium (VI) is considered much more toxic that chromium (III) as both the inhalation and ingestion can cause gastrointestinal problems including abdominal pain, vomiting, and hemorrhage.

Long-term health effects: Long-term effects of chromium (VI) inhalation are on the respiratory tract that causes perforations and ulcerations of the septum, bronchitis, and pneumonia. Chromium exposure may cause effects on the liver, kidney, gastrointestinal and immune systems.

Cancer Risk: Chromium (VI) is categorized by the EPA as a Group A, known human carcinogen by inhalation route.

Concentration of arsenic (As):

Arsenic is released in larger quantities in the environment during volcanic eruptions, rock erosion, forest fires, and many human activities. The wood preserving industries are using about 90% of the industrial arsenic. It is widely used in the manufacture of paints, dyes, drugs, soaps and semi-conductors.

Ambient air concentration: Arsenic concentrations measured in thirteen cities across the U.S. in air were 2 ng/m^3 (Chen and Lippmann 2009). The ToxGuide for arsenic lists environmental levels in air ranging from 1 to 6 ng/m^3 in remote locations and 20-100 ng/m^3 in urban areas (ATSDR, 1994). The concentrations of arsenic were below safe limits at different locations as shown in the figure 4.19-4.21. The study reveals that the city is not contaminated with this metal and the mitigation methods for this metal in the given areas are not required but continuous evaluation is needed to keep check on the existing conditions.

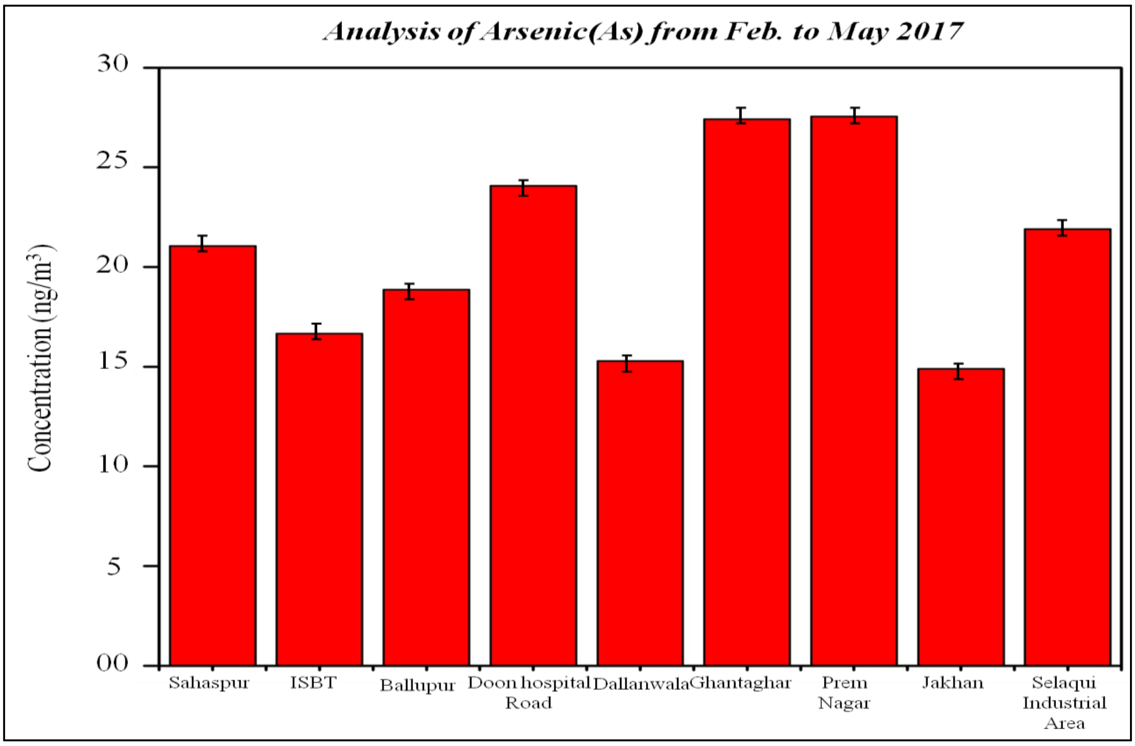

Figure 4. 19 Concentration of arsenic at different locations from Feb. to May 2017.

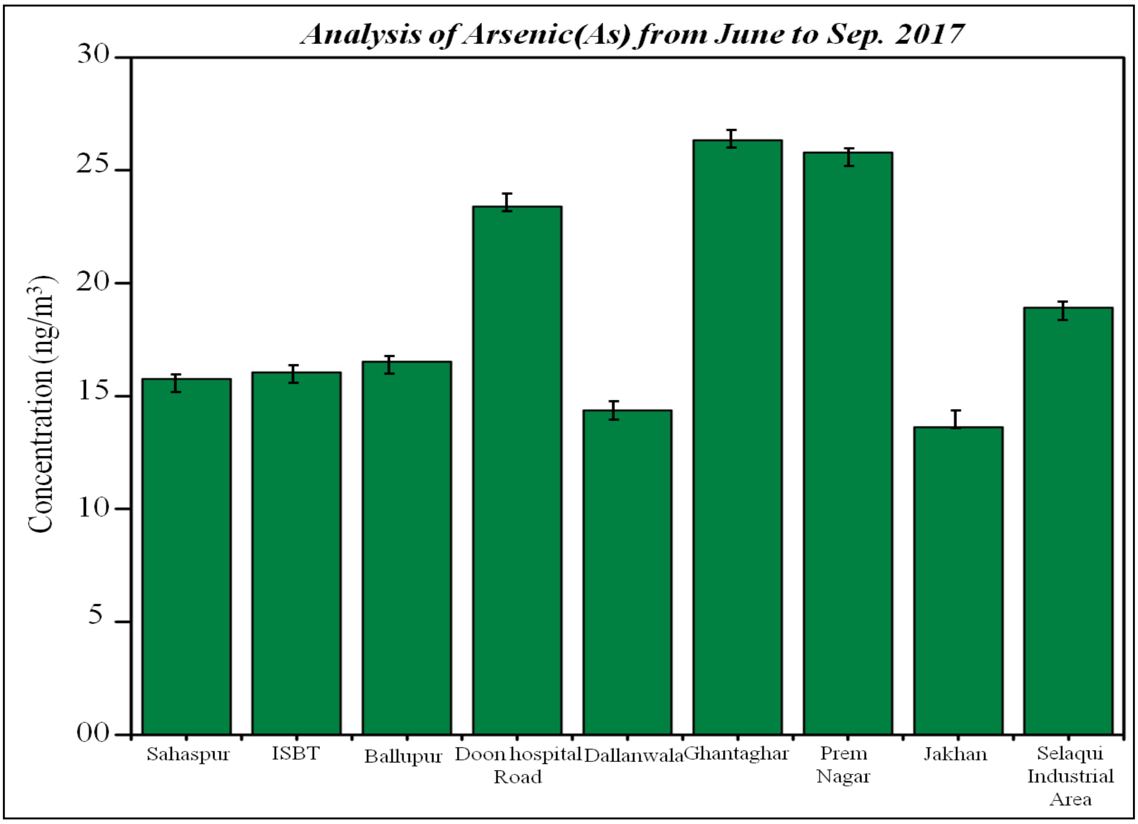

Figure 4. 20 Concentration of arsenic at different locations from June to Sep. 2017.

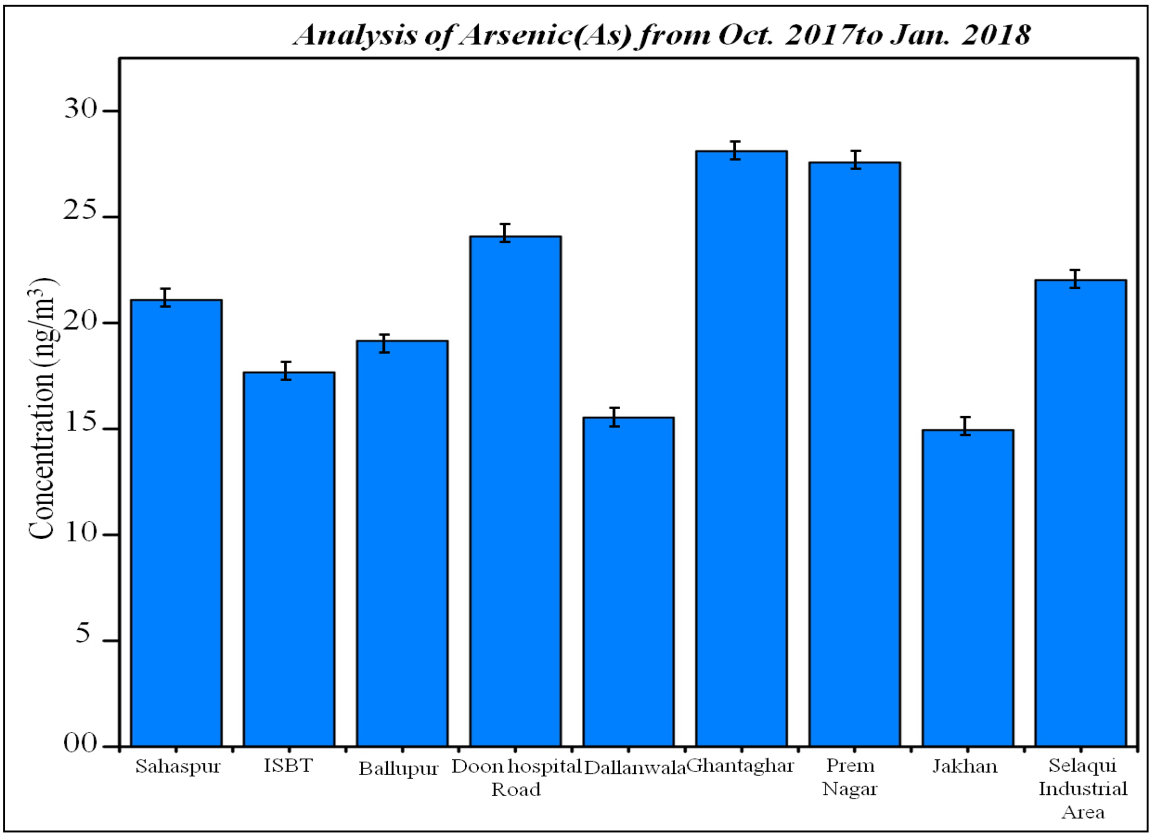

Figure 4. 21 Concentration of arsenic at different locations from Oct. 2017 to Jan. 2018.

Short-term health effects: The short term exposure to arsenic may cause nausea, diarrhea, and abdominal pain. In addition, acute inhalation can lead to central and peripheral nervous system disorders. Acute levels of arsine can cause death, while moderate amount exposure decreases the production of RBCs and WBCs, abnormal cardiac rhythm, and an unusual sensation in hands and feet (Bissen and Frimmel, 2003).

Long-term health effects: Inhaling arsenic can cause skin and irritation on the mucous membrane. Low level exposure if happening for longer duration can cause skin darkening and small corns or warts on the palms, soles, and torso.

Cancer Risk: As per the EPA, it is a Group A human carcinogen and can be the cause of cancer of the skin, lungs, liver, and bladder.

Concentration of selenium (Se):

Selenium is widely used in the electronics industry, glass industry, drugs preparation, anti-dandruff shampoos, formulations of fungicides and pesticides. It is also utilized as an additive for a nutritional feed of poultry and livestock.

Ambient air concentration: An average ambient air selenium concentration is estimated to be below 10 ng/m^3. Concentration ranges of selenium associated with particulate matter in urban atmospheres were 0.2 to 30 ng/m^3 in the U.S. (HSDB, 2010). The selenium concentrations were below permissible limit at all the chosen sites of study except at Ghantaghar area and Prem Nagar area where the concentration was found little elevated in all the seasons as shown in figure 4.22-4.24. Therefore, the areas where the concentration of the selenium was elevated it is suggested that proper mitigation methods should be employed to bring down the level and lower down its health effects on the residents. Furthermore, various government agencies may issue advisories to take precautions while they travel to those areas.

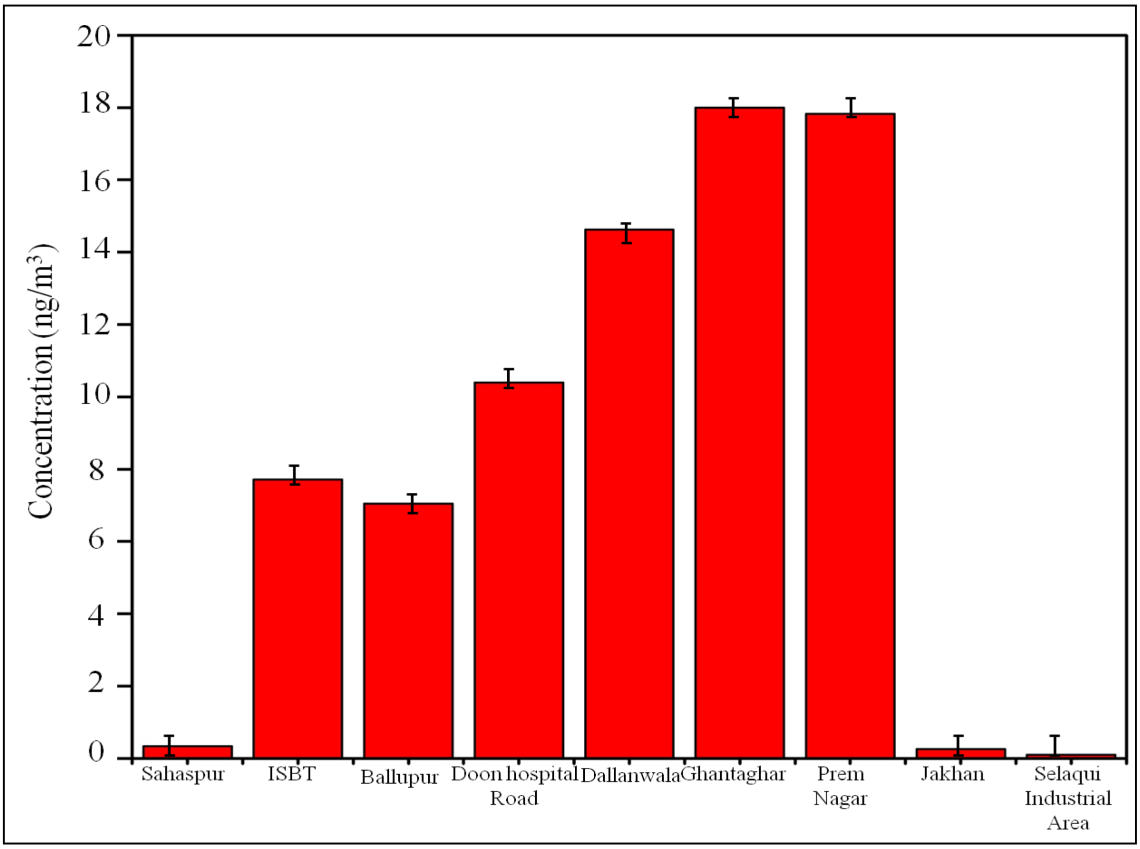

Figure 4. 22 Concentration of selenium at different locations from Feb. to May 2017.

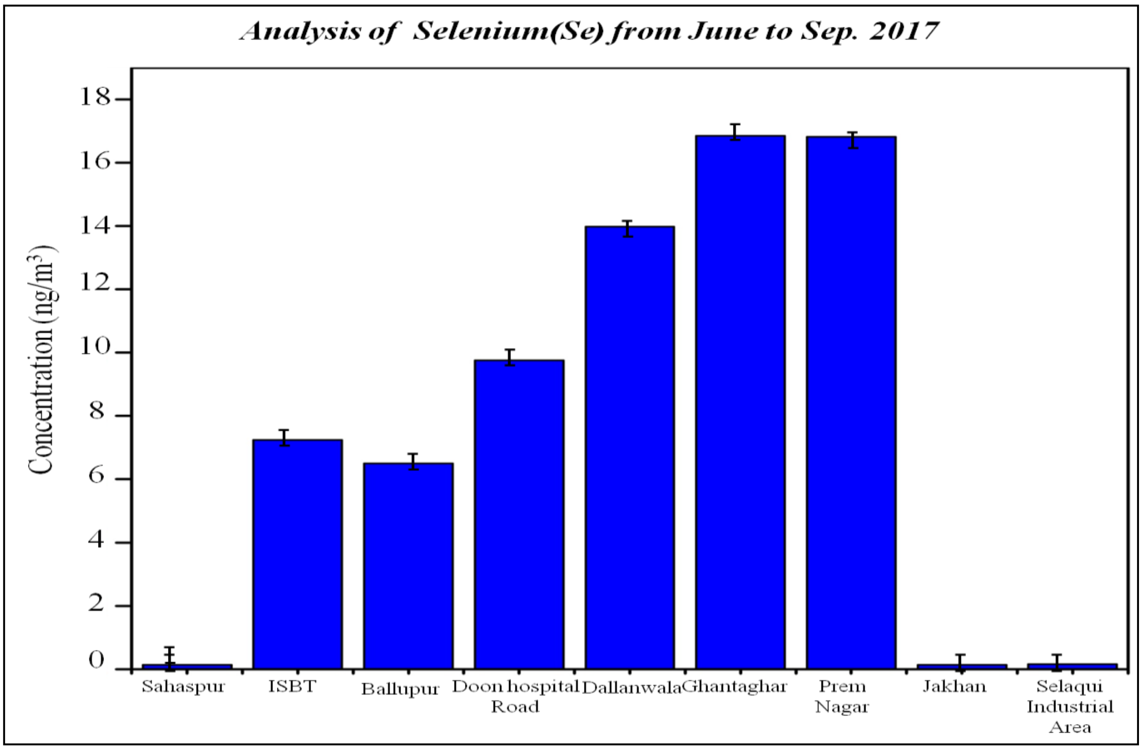

Figure 4. 23 Concentration of selenium at different locations from June to Sep. 2017.

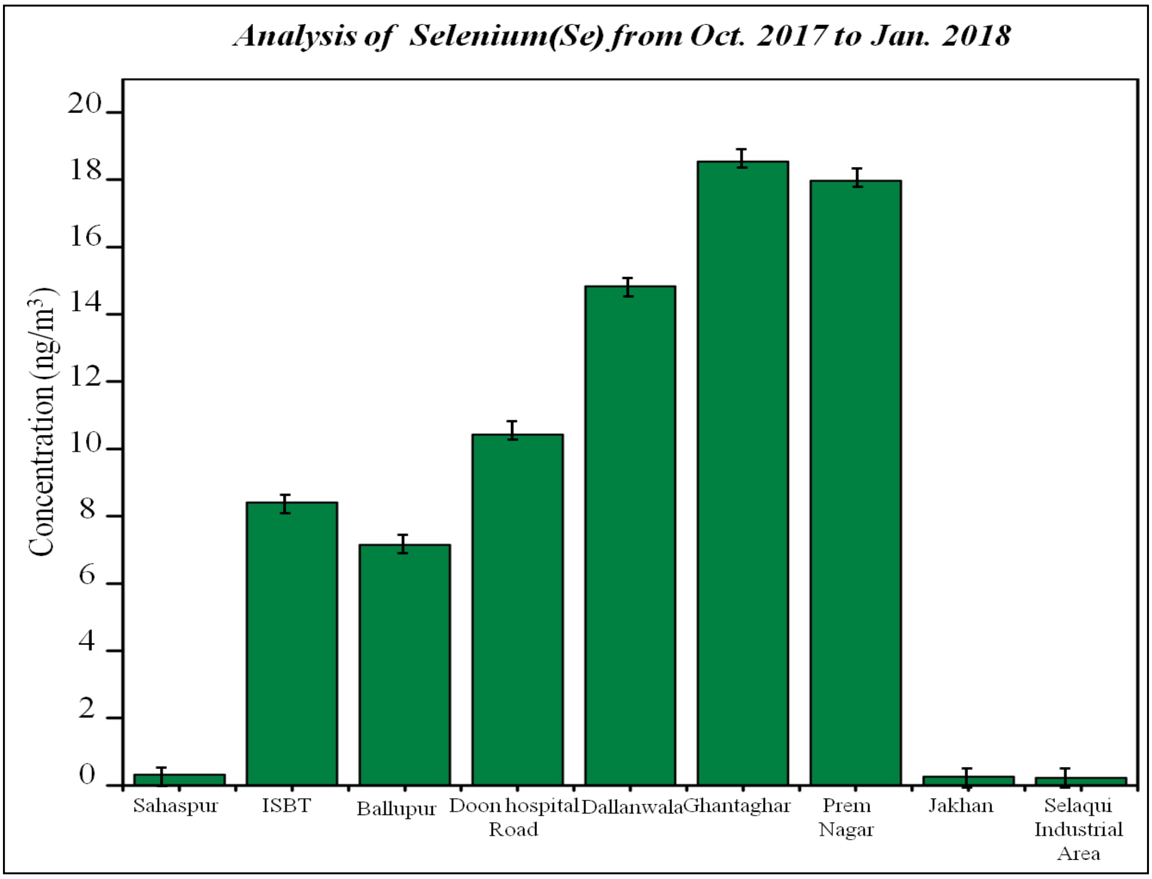

Figure 4. 24 Concentration of selenium at different locations from Oct. 2017 to Jan. 2018.

Short-term health effects: Acute exposure to selenium compounds for example hydrogen selenide, selenium dioxide can cause respiratory problems. Short-term exposure to elemental selenium dust can cause problems on the mucous membranes of the nose and throat, may cause nosebleeds, bronchial spasms, bronchitis, and chemical pneumonia (Vinceti et. al. 2001).

Long-term Health Effects: Long-term exposure to high selenium levels via ingestion causes skin discoloration, loss of hair, nails and tooth.

Cancer Risk: According to the EPA the elemental selenium is a Group D, not classifiable as to human carcinogenicity.

The data collected from different sites in Dehradun for the eight heavy metals presence in the air in the study clearly demonstrate that some areas in Dehradun are polluted with these metals when compared with the different the prescribed concentration limits set by different environmental agencies nationwide but not to the alarming level. In some areas such as Ghantaghar, Premnagar, Sahaspur, Dallanwala and others the concentration is rising due to the vehicular pollution, industrial activity and cutting of trees for buildings. It is predicted from the study that if the level rises to greater extent, many health related problems will arise in the residents of the area. Furthermore, it is also suggested that some mitigation methods which are employed in different countries for lowering down the air pollutants should be employed to a greater extent to stop further deterioration of the air.

Mitigation methods:

In order to mitigate or reduce the exposure of heavy metals, risk analysis should consider not only the direct emissions to the atmosphere but also the impacts of heavy metals deposition and transfer through the food chain and drinking water. Therefore, complete exposure assessment is required. Apart from this, cost-benefit analysis for measures should also be assessed based on the impacts resulting from the exposure.

At the individual level, the measures include:

(i) Limit the exposure.

(ii) Decrease absorption with dietary supplements that will inhibit intake and uptake of cadmium, mercury and lead.

It has been found that Fe reduces uptake of Cd and Pb; Ca reduces the uptake of Cd; high sulphur foods such as onions and garlic probably help the body blocking uptake and decreasing retention of many toxic metals, thus decreasing the release of stored metals and protecting the vulnerable tissues (VMICC, 2003).

While general population can contribute to heavy metals mitigation process by stop buying metal free products for example, Hg. Furthermore, correct disposal of the products containing heavy metals and reduction in the demand for products causing harm to the environment (EPA, 2009).

A European database has suggested methods and options for reducing the metals occurrence in Europe from industrial facilities. Experts of different countries that were involved in the process of the data collection (ESPREME, 2004). The general Access database contains 483 files referring to the processes responsible for emissions to the air, the measures taken for controlling these emissions, the investment and operating costs for each process, the unabated emission factors generated from individual process and the implementation of specific abatement methods.

General options for reducing emissions of heavy metals and their compounds

Emission reduction measures mainly focussed on add-on technologies and process modifications that include maintenance and operating control. The following measures are available:

(a) Application of low-emission process technologies in all new installations.

(b) Off-gas cleaning (secondary reduction measures) with filters, scrubbers, absorbers and others.

(c) Change or preparation of raw materials, fuels and/or other feed materials (e.g., use of raw materials with low heavy metal content).

(d) Changes in management practices such as good housekeeping, preventive maintenance programmes, or primary measures such as the enclosure of dust-creating units.

(e) Appropriate environmental management techniques for the use and disposal of certain products containing Cd, Pb and/or Hg.

Control techniques

- The control techniques that are available for the Cd, Pb and Hg emission abatement are base measures that include raw material and/or fuel substitution and low emission technologies, and the other measures, such as fugitive emissions control and off-gas cleaning.
- In the case of particle-bound emissions of Cd, Pb and Hg, the metals can be captured by dust-cleaning devices.
- A major source of anthropogenic mercury emissions is the combustion of coal in utility and industrial boilers. The heavy metal content is very high coal than in oil or natural gas. Hence, it is advisable to switch fuel if fuels with lower mercury

content are available for example natural gas or specific types of coal having less mercury content.

- Integrated gasification combined-cycle (IGCC) power plant technology is a high efficiency technology that can have reduced emissions compared to large scale power production based on solid fuels that do not use IGCC.

- To reduce the emissions of heavy metals is generally the application of high performance dedusting devices such as electrostatic precipitators (ESP) or fabric filters (FF) (ESPREME, 2004).
- Particulate control techniques (electrostatic precipitators, filters, cyclones, wet scrubbers).

- Pulse Jet Dust Collections Systems - Pulse jet filtration has become the preferred choice all around the world. Researchers are continuously striving for new concepts of more energy-efficient and compact particulate collector which can meet future emission limits and operating requirements. The system can also satisfy stringent emission norms and if required, it can be embedded with new technique for simultaneous control of particulate and gaseous pollutants.
- Adsorption methods (activated carbon, impregnated sorbents, etc.).

Municipal, medical and hazardous waste incineration:

Some wastes can neither be classified as hazardous, municipal nor medical waste, that may be incinerated and hence, potentially constitute a source of heavy metal emissions. Additionally, there may be some thermal waste treatment methods that may also become the source of heavy metal emissions.

Generally the emissions caused of the cadmium, lead and mercury are the result of incineration of municipal, medical and hazardous waste. Measures should be taken both before and after incineration so that these emissions can be reduced to zero. Primary techniques for preventing the emissions of mercury into the air before incinerating are those that prevent or control the inclusion of mercury in waste. In some countries, mercury-containing components are collected separately from the waste stream so that they can be managed or recycled properly. Removal of mercury items from the waste stream before it enters the incinerator is highly cost-effective process than capturing mercury later using emissions control devices.

The best technique for dedusting and reducing heavy metals emissions is FFs in combination with both dry and wet methods for controlling volatiles. ESPs in combination with wet systems can also be designed to reach low dust emissions, however they offer fewer chances than FFs, especially with pre-coating for adsorption of volatile pollutants.

The Peck process is also a very promising technique with reduced heavy metals emissions in the flue gas. It has been developed for municipal solid waste treatment but can be applied to other wastes. The other options to reduce heavy metals emissions in the air are the heavy metal evaporation process and the hydro-metallurgical treatment plus vitrification.

Phytoremediation:

Mitigation of air pollution is emerging as one of the biggest challenge for the environmental management agencies all around the world. Hence, the use of plants for the reduction of air pollution is now being given priority. It has been found plants play an important role in filtering ambient air by adsorbing particulate matter onto leaf surfaces and thus, they act as a sink for both particulates and gaseous pollutants. Lohr & Pearson-Mims (1996) reported that plant canopies help in reducing atmospheric particle concentration. Furthermore, in some studies it has been estimated that increasing tree cover could reduce the concentration of PM_{10} in the air by 26%, causing the removal of about 200 tons of PM_{10} per year (McDonald et al. 2007). Nowak (1994) elucidated that exposed surfaces of plants act as persistent absorbers of pollutants in the environment for example, in a single season, trees in

Chicago USA removed 212 tons of PM_{10} from the air and they can remove 711,000 metric tons of air pollutants from the cities (Nowak et al. 2006). It is also reported in some studies that leaves act as an efficient filters of airborne particles due to their large size and high surface to volume ratio. In addition to this, foliar features such as roughness of leaf surface, frequency & length of trichomes and size & frequency of stomata also play important role in determining the efficiency of plants to trap dust particulate (Neinhuis & Barthlott 1998). Plants differ considerably in terms of their ability to trap dust because of the differences in their leaf surface characteristics like epicuticular wax, cuticle, epidermis, stomata and the trichomes. Hence, selection of appropriate type of plant that is embedded with all suitable morphological features to capture PM and tolerance of stress are both important traits in the selection of optimal vegetation for urban and suburban areas for air pollution abatement strategy.

Plants used for mitigating air pollution:

Some of the important and widely available are neem (*Azadirachta indica*), peepal (*Ficus religiosa*), banyan (*Ficus benghalensis*), almond (*Terminalia catapa*) which serve as excellent quantitative and qualitative indices of pollution level (Wagh et al. 2006). It has been found that the most efficient shrub species examples for the removal of PM are *Stephanandra incise*, *Pinus mugo*, *Skimmia japonica*, *Spiraea japonica* and *Syringa meyeri* as shown in figure 4.25 (Popek et al. 2013; Sæbø et al. 2012; Dzierzanowski et al. 2011).

Trees that are proven to be effective for the reduction of PM from the air and are suggested for setting up green belts for containment of air pollution in the human environment, especially in industrial and urban areas, are Ficus carica, Toona ciliate, Betula pendula and many more (Sæbø et al. 2012). Unfortunately, in the temperate climate, most of these plants shed leaves for winter. Therefore, in countries where the growing season is short and high concentrations of PM are emitted in the winter season, evergreen species could be a more suitable choice for urban plantings.

Figure 4. 25 Shrubs (a) *Pinus mugo* (b) *Skimmia japonica.* Image adapted from *www.thespruce.com* and *www.lovethegarden.com*

The efficiency of evergreen species in PM accumulation has been described in several studies (Mori et al. 2016; Przybysz et al. 2014; Sæbø et al. 2012). Among the evergreens, coniferous plants are an excellent choice for air purification due to the abundant wax layer on the needles, smaller leaves, and more complex shoot structures (Freer-Smith et al. 2005). These features, along with conditions causing high air turbulence inside the tree crowns, contribute to higher interception capacity of contaminants by coniferous plants. Moreover, evergreen conifers have the potential for accumulating toxic pollutants throughout the year as most of these plants keep their needles for several years there is no possibility of recycling PM accumulated on needles every year (Gawroński et al. 2017). Therefore, conifers such as *Taxus baccata*, *Pinus sylvestris*, evergreen broad-leaved plant *Hedera helix* as shown in figure or plants which keep last year foliage through the winter period *Carpinus betulus* are found to be most effective in the abatement of PM from the air (Przybysz et al. 2019).

Figure 4. 26 Evergreen plants (a) *Taxus baccata* (b) *Hedera helix.* Image adapted from www.123rf.com and powo.science.kew.org.

Plant canopies provide a naturally absorbing surface with a favourable surface area/volume ratio and a long life-span. Leaves have been shown to be able to capture coarse, fine and ultra-fine particles, including transfer of ultra-fine particles to the inner layer by diffusion. Recently, it has been investigated by some researchers that the most common pollutants associated with fine particles in contaminated atmosphere are trace elements especially heavy metals. A significant burden of heavy metals accumulated on vegetative surfaces has been demonstrated by trace element investigations conducted in roadsides, industrial and urban environments (Rossini Oliva & Rautio, 2004; Rossini Oliva & Valdes, 2004). Datta & Ghosh (1985) reported the presence of high lead content in banyan leaves exposed to vehicular exhaust. Concentration of different heavy metal particles Al, Ba, Cr, Cu, Fe, Pb, Mg, Mn, and Zn were determined in leaf samples of *N. oleander* by Mingorance & Oliva, 2006. They carried out multivariate analysis classifying the sampling sites in four groups based on the metal content in vegetal leaves in agreement with traffic and human activity site. It was found that about 30% of Al, Fe Cr, Cu and Pb originated from aerial deposition on leaves.

Phylloremediation:

Phylloremediation is a natural and environmentally friendly way of bioremediation of air contaminants. This is a process in which plant leaves and leaf-associated microbes are used for the remediation of air pollutants as shown in figure 4.27.

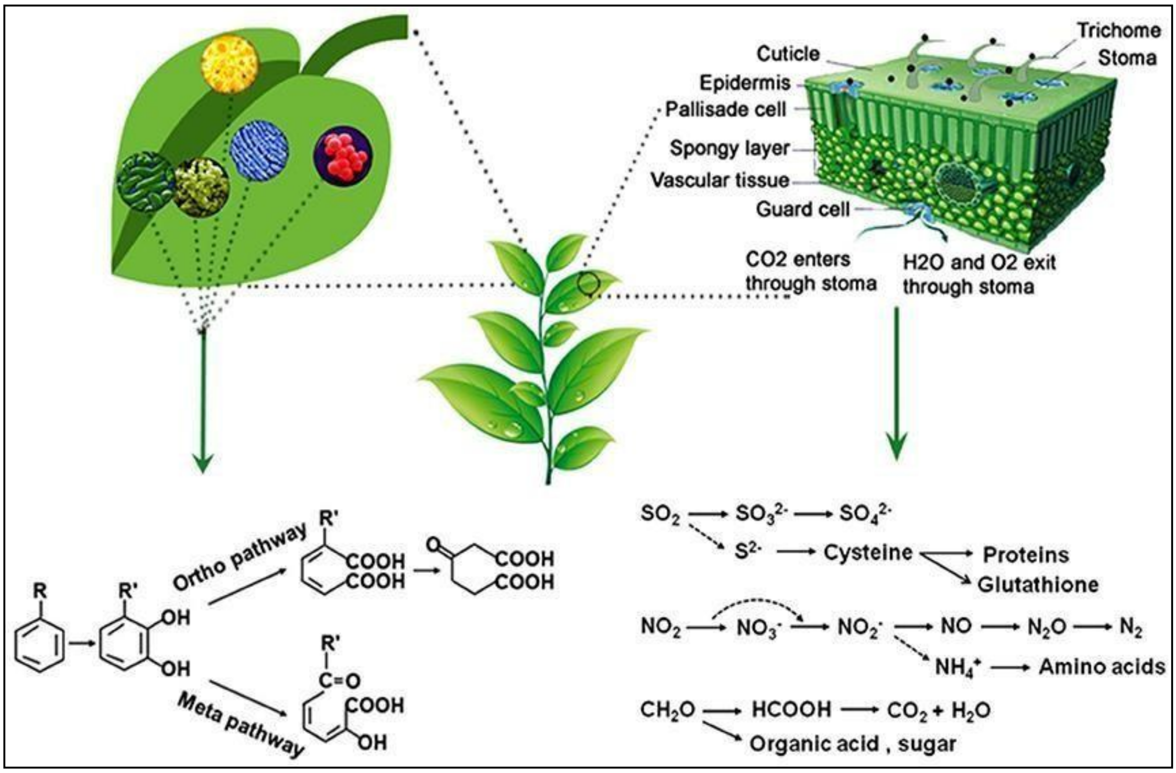

Figure 4. 27 A schematic illustration of phyllosphere. Reprinted with permission from Ref. Wei et. al. © Frontiers 2017

The main component of this technology is present in plants which can adsorb or absorb pollutants and they also support microbes in biodegradation or biotransformation of pollutants (Wei et al. 2017).

Many researchers have documented the plant responses to pollutants such as NOx, SO2, O3, and VOCs. For example, De Kempeneer et al. (2004) found that azalea leaves and the leaf-associated Pseudomonas putida in reducing VOCs, leaves of yellow lupine plants along with endophytic Burkholderia cepacia for toluene reduction (Barac et al. 2004), and poplar leaves and the leaf-associated Methylobacterium sp. decreased xenobiotic compounds (Van Aken et al. 2004).

---End of Chapter 4---

CHAPTER-5

CONCLUSION

CHAPTER 5

CONCLUSION

As the new state of Uttarakhand was formulated on the year 2000, its capital city, Dehradun, has suffered tremendous pressure because of the increasing population and more number of vehicles. The pollutants get added up in the environment through both the natural sources as well as anthropogenic sources. The air pollutant generally includes particulate matter, harmful gases, and heavy metals. In the study we have obtained the data from different locations of Dehradun which firmly illustrates that some of the parameters (Selenium, Lead, Arsenic, copper) have crossed the permissible safe limits set by the OSHA and USEPA which are considered unsafe for human inhabitation. While the Heavy metals are on rise at Ghantaghar, Selaqui, Sahaspur and premnagar areas and crossing the prescribed limits of OSHA, USEPA and CPCB. Ghantaghar area is the oldest inhabited market place of Dehradun and the centre point, making it the most crowded place in Dehradun which clarifies the significant RSPM concentration and slight increasing trend of the concentrations of heavy metals in the ambient air quality. Premnagar is primarily residential area where a sudden rise in concentration of heavy metals can be observed. Since Selaqui and sahaspur is considered to be an industrial area so the concentration of heavy metals is on the higher side. Almost all the major inhabited areas of Dehradun are located near the surroundings of these nine locations. Hence, it can be concluded that the Air quality of Dehradun city is deteriorating of an alarming situation and may pose threat to health conditions of certain special groups of the society like infants, children and aged people.

The high concentration of pollutions was found in various sampling location which was mainly due to vehicular pollution in Dehradun city and due increased urbanization and industrialization over the past decade.

The different mitigation methods can be employed for the assessment of the sources and emissions to environment in the city. Also, the emission control with more reliable equipment may result in more efficient regulatory efforts to reduce heavy metal emissions from anthropogenic sources and it is clear that the most effective way to reduce or to eliminate the exposure to heavy metals in the environment is taking actions at the sources level (EEA, 2010).

For the effective management of air pollution huge importance must be given towards the identification of the sources responsible for the addition of the pollutants in the ambient air. After identification it is also critical to foresee the situation and employ suitable methods to mitigate the problem so as to prevent threats and risks before they become problem to human health.

---End of Chapter 5---

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- Srivastava, A.,Siddiqui, N.A., Koshe k Rupesh 2016, November. Human Health Effects Emanting from Airborne Heavy Metals Due to Natural and Anthropogenic Activities: A Review. *International journal of civil and environmental engineering. Pp 279-305*
- Srivastava, A., Mondal, A., Siddiqui, N.A., Tauseef, S.M., 2020, Analysis and Quantification of Airborne Heavy Metals and RSPMs of Dehradun City. *Nature Environment and Pollution Technology. P-ISSN- 09726268 vol.19.*

SCHOLAR'S PROFILE

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01/09/2020 - Till Now

Publications

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