Source Identification of Atmospheric Particles Deposition in Urban and Rural Areas of Assam

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DOCTOR OF PHILOSOPHY

by

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DECLARATION

I hereby declare that except where specific reference is made to the work of others, the content of this dissertation are original and have not been submitted in whole or in part for consideration for any other degree or qualification at this or any other country. I do also declare that the matter embodied in this thesis is the result of investigations carried out by me in the Department of Civil Engineering, Indian Institute of Technology Guwahati, Guwahati, Assam, India.

In keeping with the general practice of reporting scientific observations, due acknowledgements have been made wherever the work described is based on findings of other investigators.

Place: IIT Guwahati

Date: 11-02-2020 **Rajyalakshmi Garaga**


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CERTIFICATE

This is to certify that the thesis entitled "**Source Identification of Atmospheric Particles Deposition in Urban and Rural Areas of Assam**", submitted by **Rajyalakshmi Garaga** (Roll No. 156104006), to the Indian Institute of Technology Guwahati, for the award of degree of Doctor of Philosophy in Civil Engineering, is a record of bonafide research work carried out by her under my supervision and guidance. The thesis work, in my opinion, has reached the requisite standard fulfilling the requirement for the degree of Doctor of Philosophy.

The results contained in this thesis have not been submitted in part or full to any other University or Institute for award of any degree or diploma.

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To the God
who is,
who was and
who is to come

-from Bible

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ABSTRACT

India, the 2nd most populous country, is identified as the 11th mega biodiversity center in the world and 3rd in Asia. The country shares approximately 4.2% of total geographical area for vast conservation of natural ecosystems. However, the increasing anthropogenic activities like expansion of industries, urbanization and huge consumption of fossil fuels led to detrimental effects on varieties of ecosystems and deterioration of air quality. According to world health organization (WHO, 2016) report, out of world's 20 most polluted cities, 14 cities belonging to India pose major environmental risk due to air pollution to human health.

Numerous epidemiological and toxicological studies have insisted an association between particulate matter (PM) and reduced life expectancy, high mortality and morbidity rates. However, the effect of PM on human health is directly linked to the size of the PM which can deeply penetrate into the human respiratory tract. The direct and indirect effects of PM is largely monitored not only by its size, but also by its chemical composition and concentration. Eventhough studies related to total mass of PM are increasing, there are very few studies in India focusing on the size segregated PM. Also, in countries like India, having the world's highly polluted cities, wet deposition can provide great relief in subsiding the air pollution. However, the chemical composition, especially Sulfur and Nitrogen compounds of the rain water could be of particular concern. Thus, a combined analysis of both wet and dry depositions and the systematic mechanism of these processes should be of prime focus.

Therefore, the present study was conducted in Assam, which is identified as one of the 200 eco-regions in the world and the most populous state of northeast India. A total of five locations (three urban (S1, S2, S3) and two rural (S4, S5)) for size resolved aerosol dry deposition and one urban location for wet deposition were considered for the study. Laboratory testing techniques were adopted to comprehensively characterize the samples collected. The regional deposition of these measured elements of size resolved PM was evaluated in order to understand their effects on human respiratory tract. Also, the data of Indian air quality studies from literature was collated and compared with the present study. In addition, source apportionment analysis using the US EPA's Positive Matrix Factorization (PMF) model was performed to assess the dominant sources and their contributions for both wet and dry depositions.

Results obtained from each of these deposition mechanisms were analyzed independently and discussions were presented for both wet and dry separately. Out of 40 rain events sampled, 31 were acid rain events with pH <5.6. The occurrence of acid rain events in this region, was elaborately discussed in terms of chemical characterization and neutralization factor. Dominance of SO_4^{2-} (29%) and NO_3^- (27%) and poor neutralizing capacity of Ca^{2+} (0.09) and Mg^{2+} (0.001) could be the reason for acid rain in this region. Isotope analysis along with the back trajectories could identify the origin of rainwater droplet during monsoon and non-monsoon seasons. The results obtained were further supported by the source identification method using US EPA's PMF. The wet deposition analysis in this study could distinguish the sources of moisture during monsoon as continental origin with marine source (40%) as dominant. While, in the case of non-monsoon the origin of rain droplet was from water in-land with greater contributions from industrial sources (28%).

The size segregated samples collected in the selected locations were carefully analyzed and discussed in terms of regional deposition in human respiratory tract using inhalation and deposition curves. The concentrations of PM_{10} and $PM_{2.5}$ in the five sites exceeded the prescribed Central Pollution Control Board (CPCB) standards. Seasonal variation of

fractional deposition of PM in human respiratory tract was observed. For example, during winter, in one of the urban sites i.e. S3 (0.61) the maximum deposition was in Pulmonary (P) region, while in the case of other sites, the maximum deposition was in Nasopharyngeal (NOPL) region. Analytical formulations have been proposed based on regression analysis of the experimental data. PMF revealed five to eight factors at each individual site in NOPL, TB and P regions: biomass burning (accounting for 7-32% of PM), coal combustion (14-27%), construction dust (9-25%), dust emissions (17-28%), industrial emissions (12-26%), oil refinery (18%), secondary aerosols (17-33%) and vehicular emissions (12-39%). Clear distinction between urban and rural environments was observed with dominant sources as vehicular emissions in urban and biomass burning and dust emissions in rural areas. Finally, the results of both wet and dry depositions were combined to provide comprehensive understanding of dominant sources in this region. The common and dominant sources in wet and dry depositions were dust emissions (26% and 29%) and vehicular emissions (18% and 19%). Therefore, this combined approach of both wet and dry depositions forms a basis upon which the future air quality studies can be formulated in this region and also facilitates the findings to be utilized to design better air pollution mitigation strategies in this region.

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NOTATIONS AND ABBREVIATIONS

δD	Hydrogen isotope
$\delta^{18}O$	Oxygen isotope
d-excess	Deuterium excess
X_{ij}	Speciated concentration
e_{ij}	Residual
UC_{ij}	Uncertainties
Q	Object function
AAS	Atomic absorption spectrometry
AEP	Acute eosinophilic pneumonia
AGL	Above ground level
ARL	Air Resource Laboratory
ARI	Acute respiratory infections
AT	Averaging time
ASI	Automatic Sample Injector
BAPMoN	Background Air Pollution Monitoring Network
CBHI	Central bureau of health intelligence
CFC	Chlorofluorocarbons
CMB	Chemical mass balance
C-NES	Centre for northeast studies and policy research
CPCB	Central pollution control board
DNA	Deoxyribonucleic acid
EANET	Acid deposition monitoring network in East Asia
EC	Elemental carbon
ED	Exposure duration
EF	Enrichment factor
EF	Exposure frequency
FA	Factor analysis
GAW	Global Atmospheric Watch
GDAS	Global Data Assimilation System
GMWL	Global meteoric water line

HI	Hazard Index
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory
IC	Ion chromatography
IGP	Liquid petroleum gas
IITM	Indian Institute of Tropical Meteorology
IMPROVE	Interagency Monitoring of Protected Visual Environment
Km	Kilometre
LGR	Los Gatos Research
LMWL	Local meteoric water line
LPG	Interagency Monitoring of Protected Visual Environment
M	Concentration of metal
MDL	Method detection limit
MLR	Multiple linear regression
NA	Not available
NAAQS	National ambient air quality standards
NCAP	National Clean Air Programme
NF	Neutralization factor
NH	National highway
NHAI	National highways authority of India
NHP	National health profile
NIST	National Institute of Standards & Technology
NOAA	National Oceanic and Air Administration
NOPL	Nasopharyngeal
NSS	Non sea salt
OC	Organic carbon
P	Pulmonary
PAHs	Poly aromatic hydrocarbons
PAN	Peroxyacetyl nitrate
PCA	Principle component analysis
PM	Particulate matter
PM_{10}	Particles less than aerodynamic diameter of 10 µm
$PM_{2.5}$	Particles less than aerodynamic diameter of 2.5 µm
$PM_{2.1}$	Particles less than aerodynamic diameter of 2.1 µm

PMF	Positive matrix factorization
PTFE	Polytetrafluoroethylene
RFC	Reference concentration
RAIS	Risk Assessment Information System
S/N	Signal to noise ratio
S1	Jalukbari
S2	Noonmati
S3	ABC Tarun Nagar
S4	Barpeta
S5	Jorhat
SEARCH	Southeastern Aerosol Research and Characterization
SRM	Standard Reference Material
STN	Speciation Trends Network
TB	Tracheobronchial
TOC	Total organic carbon
TSP	Total suspended particles
UNMIX	Unmix
UNESCO	United Nations educational, scientific and cultural organization
USEPA	United States environmental protection agency
VOCs	Volatile organic compounds
WHO	World health organization

CHAPTER I – INTRODUCTION

1.1 PREAMBLE

Atmospheric deposition is generally considered as major pathway through which the substances from atmosphere can enter the terrestrial and aquatic ecosystems. It is a source of all organic and inorganic inputs in to the atmosphere, such as dusts, acids, metals, nutrients and pollutants (Weyhenmeyer et al., 2009). Excessive presence of these substances in the atmosphere that can adversely affect the human health and environment is basically termed as air pollution (Kampa and Castanas, 2008). However, these pollutants which are produced by different sources are transported in to air and undergo transformation. Further, they reach back to earth causing various adverse effects on diverse ecosystems (Fig 1.1). Thus, this entire process operates in a cyclic manner (Dasch, 1985).

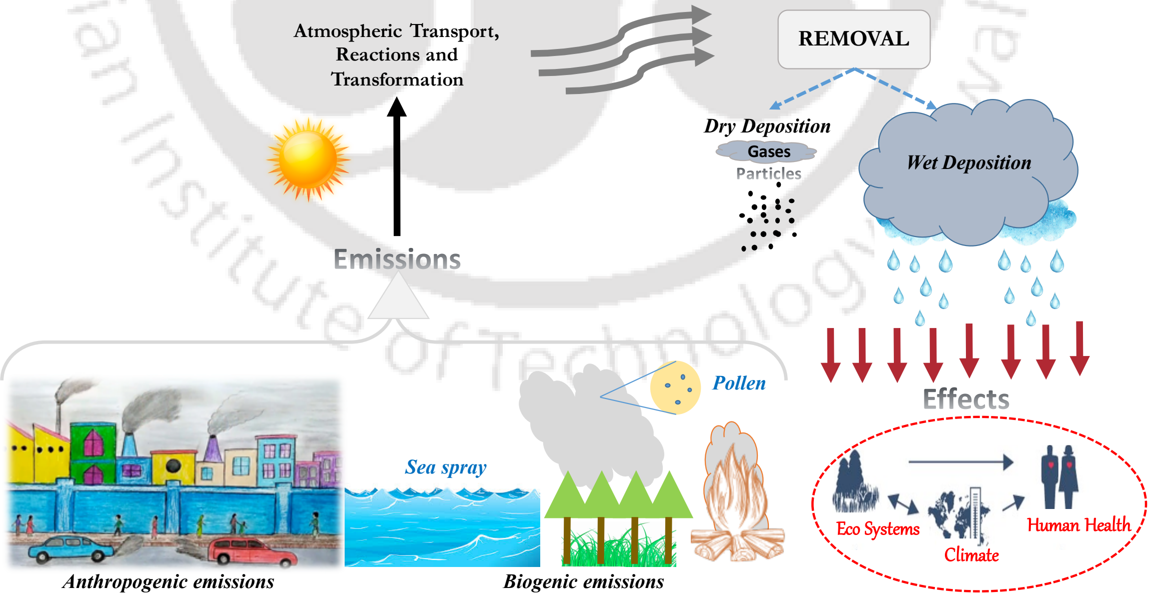

Fig 1.1 Cyclic process of pollutants in the atmosphere

The sources of air borne particles causing air pollution can widely range from natural, such as volcanic activities to larger volume of emissions from anthropogenic activities with varied effects (Table 1.1). The mode of emission by which the particles are induced in to the atmosphere can be direct (i.e. primary aerosols) or indirect resulting physical and chemical transformation within the atmosphere (i.e. secondary aerosols).

Table 1.1 Pollutant types, their origin and effects on human health and environment (Gheorghe and Ion, 2011)

Name of pollutants	**Origin**	**Effects**
Natural sources		
Sulfur, chlorine, and ash particulates, smoke and carbon monoxide methane volatile organic compounds (VOCs) Aerosol from deforestation and burning: CO, CO_2, NO, NO_2, N_2O, NH_4	Volcanoes, wildfires, cattle and other animals, pine trees	- Acid rain - smog - respiratory irritant - increased respiratory - diseases - damage cell membranes of plants
Anthropogenic sources		
Carbon monoxide, carbon dioxide, sulphur dioxide, nitrogen oxides, fluorides and substances with fluorine,chlorine (Cl_2), bromine (Br_2) and iodine (I_2), small dust particles, VOC, methane, ammonia and radioactive radiation.	Industry: the mining industry, oil and natural gas extraction, the energy industry based on fossil fuels - coal, oil, natural gas, the production of brick, tile, enamel frit, ceramics, and glass; the manufacture of aluminium and steel; and the production of hydrofluoric acid, phosphate chemicals and fertilizers. central heating,chemical and metallurgical industry, engineering internal combustion machinery industry, industrial waste, noises	- Respiratory irritant - acid rain - smog - increased respiratory - formation of secondary pollutants (PAN, O_3) - effect on soil fertilizer - Respiratory diseases - toxic effects on living cells - greenhouse gas effect - toxic effects - carcinogenic proprieties - accumulation in tissues - blocking of different processes - stratospheric ozone depletion
CO, CO_2, NO, NO_2, NH_3, CH_4, SO_2, oxides of heavy metals, H_2SO_4, SPM, HC, VOC, background aerosols: sea salt oxidation of sulphur containing gases, same organics, nitrous oxide (N_2O) pesticides	Agriculture: the vegetation fire, the denitrification process, in soils excessively fertilized and excessive use the pesticides , paddy field, intensive husbandry, deforestation	- Formation of secondary pollutants (PAN, O_3) - effect on soil fertilizer - respiratory diseases - greenhouse gas effect - toxic effects - acid rain - stratospheric ozone depletion
Aerosols from transport and constructions NOx, CO, HCl, Lead and other heavy metals , SPM	Motor vehicle pollution, noises	- Smog - increased respiratory diseases - damage cell membranes of plants - carcinogenic proprieties - accumulation in tissues - blocking of different processes - stratospheric ozone depletion
Domestic aerosols CFC, HC, FC, H_2S, CH_4 CO_2	Sewage plans, landfill site	- Carcinogenic proprieties - accumulation in tissues - blocking of different processes - stratospheric ozone depletion

In this modern world, deteriorating air quality has been one of the top, world's environmental issues which has gained wider attention, leading to an increase in awareness among the public. Around the globe, the resultant number of deaths due to indoor and ambient air pollution is 4.3 million and 3.7 million, respectively, of which, Asia is reported to have the death rate of 77% and 70%, respectively (Lancet, 2016). World Health Organization (WHO) states that 91% of world's population are dwelling in the places, where air quality is below the prescribed limits (WHO, 2016). Also, the recent news published by WHO suggested the number of death due to outdoor air pollution to be ~3 million worldwide (https://www.who.int/mediacentre/factsheets/fs313/en/).

Moreover, numerous studies have documented an association of particulate matter (PM) with high mortality and morbidity rates (Pope III and Dockery, 2006). The effects of PM on human health are localized to cardiovascular and respiratory systems, resulting in aggravated asthma, reduced lung functioning, premature deaths and non-fatal heart attacks (Wilson et al., 2004). However, these health effects are determined by the size of PM because of its potentiality to penetrate into human respiratory tract (Deshmukh et al., 2013a). In recent past, studies pertaining to monitoring network of dry deposition of PM in many urban and rural areas of India have been extensively increased with most of them focusing either on $PM_{2.5}$ or PM_{10}. Particles with size >2.5 µm deposit in nasal cavity or upper region of respiratory tract, whereas the particles having size <2.5 µm traverse through the tracheobronchial region reaching lungs and thereby undergoing interstitialization i.e. absorbing directly into the bloodstream, developing stringent effects (Klejnowski et al., 2012). Fig 1.2 shows the regional deposition of PM in human respiratory tract and their detrimental effects to the human health. The qualitative and quantitative effects of PM, having varied aerodynamic diameters (PM_{10}, $PM_{2.5}$, PM_1), are largely diverse. These effects predominantly depends not only on size but also on the sampling location, emission sources, meteorological factors and transformation mechanisms (Akyüz and Çabuk, 2009; Theodosi et al., 2011).

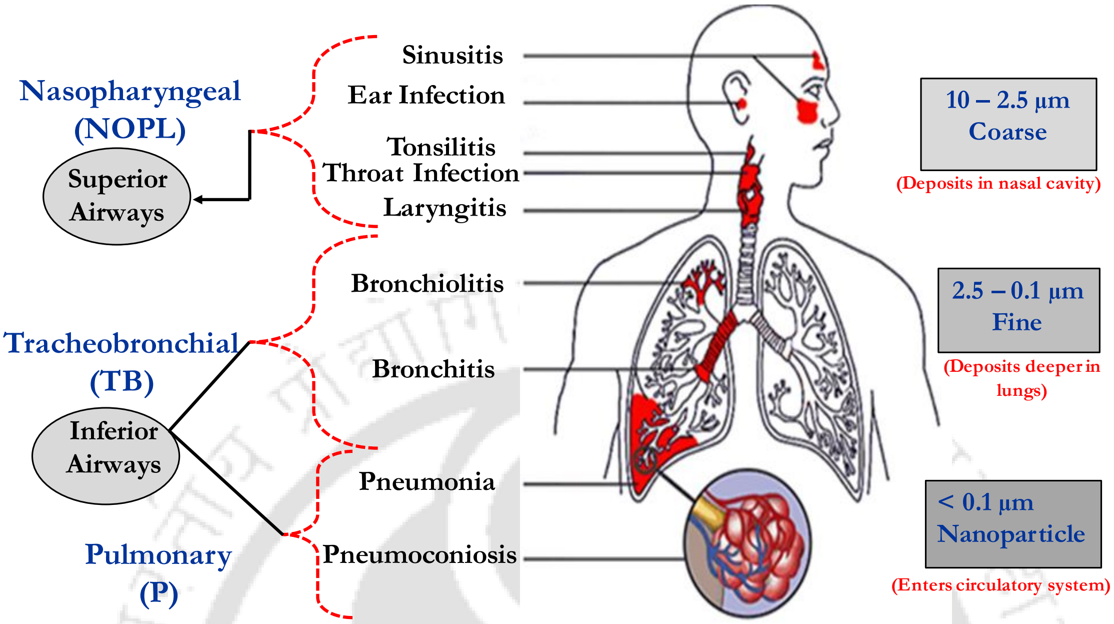

Fig 1.2 Regional deposition of relative sizes of inhaled particles in the human respiratory tract and main health effects of air pollution (after (Chang, 2010; Majkowska-Wojciechowska and Kowalski, 2012)

Atmospheric deposition, being an important means of controlling air pollution, basically comprises of both dry and wet processes as removal mechanisms (Fig 1.3). Dry deposition is referred as deposition of particles from the atmosphere through the direct transport of particle masses to the earth's surface (Dolske and Gatz, 1985). On the contrary, emissions of gaseous and particulate pollutants released into atmosphere are washed out by rains to some amount due to solubilizaton, which can cause ecological imbalance and effect in the biogeochemical cycle (Cao et al., 2009; Kulshrestha et al., 2014). In Indian scenario, where high concentrations of PM range from thoracic to alveolar size fractions, wet scavenging plays a critical role (Gobre et al., 2010; Kulshrestha et al., 1999). This process of wet scavenging often referred as wet deposition takes place in two different pathways namely below-cloud scavenging (washout) and in-cloud scavenging (rainout). Coarse particles are removed more effectively by washout process while the rainout process drains the fine particles and gases surrounding the cloud droplets present in the atmosphere (Kajino and Aikawa, 2015). Both dry and wet deposition mechanisms are strongly size dependent whose particles removal efficiency vary with different particle sizes in several orders of magnitude (Seinfeld et al., 1998). However, wet versus dry deposition depends on the availability of local precipitation. Therefore, simultaneous measurement of both wet and dry depositions in a specific location give the idea of relative and combined contributions to the total atmospheric deposition which otherwise misleads the conclusions.

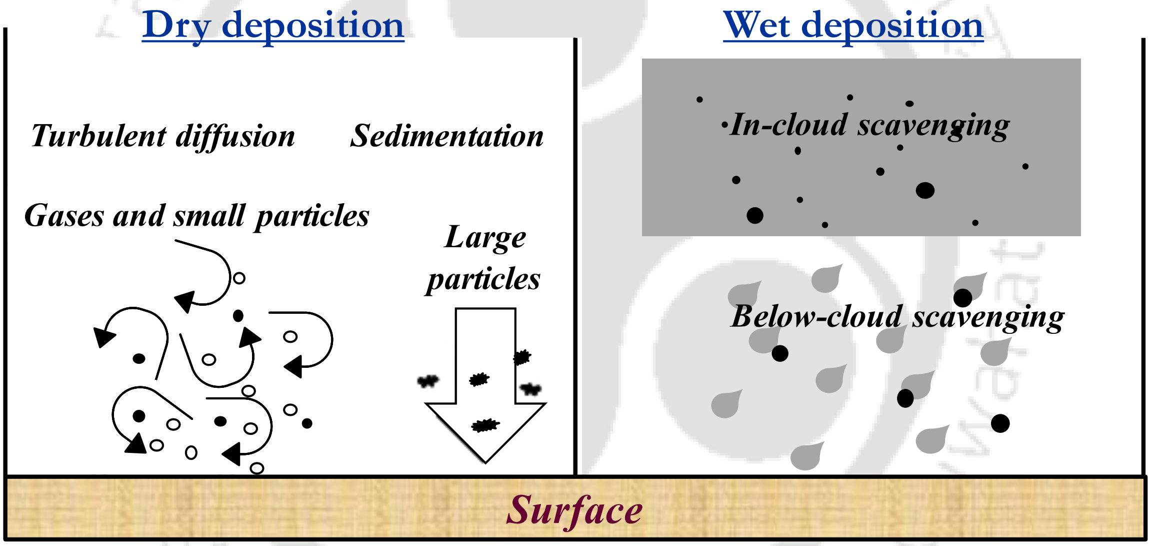

Fig 1.3 Wet and dry depositions of particulate matter in the atmosphere

1.2 NEED OF THE STUDY

Over the decades, the rapid urbanization, expansion of industrial activities, heavy consumption of fossil fuels and population explosion has led to severe air pollution at an alarming rate in world's cities (Kota et al., 2018). WHO states that, out of world's 15 most highly polluted cities, 14 cities belonging to India pose major environmental risk to human health (WHO, 2016). State of Global Air, 2019 published by Health Effects Institute (HEI), reported that India experienced 1.2 million deaths in 2017 due to outdoor and indoor air pollution (https://www.stateofglobalair.org/), claiming air pollution to be the 3rd highest cause of death among all other health risks. In this view, Indian government has initiated steps to curb the air pollution through Pradhan Mantri Ujjwala Yojana Household LPG program, Bharat Stage 6/VI clean vehicle standards, and the new National Clean Air Programme (NCAP) (http://moef.gov.in/). Current and the future strategic initiatives demand strict implementations and sustained commitments to give rise to significant health benefits in further years.

Northeast India, which is the gateway of India's variety of species is spotted as Indo-Burma biodiversity hotspot among 35 Global biodiversity hotspots (Chatterjee et al., 2006). However, the increasing anthropogenic activities like industrial revolution, huge consumption of fossil fuels and urbanization have been causing detrimental effects to the diverse ecosystems (Mehr et al., 2019; Singh and Agrawal, 2007). Modeling of global patterns for deposition fluxes of Sulfur and Nitrogen during last decade, indicated the high deposition rates in southeastern China, northeastern India, Bangladesh and central Europe (Vet et al., 2014). Such studies highlighted that the wet and dry depositions of Sulfur and Nitrogen is high in northeastern India, having good agreement between model estimates and measurements. Also, studies have suggested that acidification of Sulfur and Nitrogen have transformed 7-17% of the global area in natural ecosystems to great peril (Bouwman et al., 2002).

Assam, being the highly populated state among northeastern India is widely known for its wildlife and natural heritage. It is thus, identified as one of the 200 eco-regions in the world and recognized as center for two United Nations Educational, Scientific and Cultural Organization (UNESCO) world heritage sites (Chatterjee et al., 2006). However, the state has been affected with increasing number of acute respiratory infections (ARI) with, 20,667 ARI cases and 200 deaths (in 2016) and 22,834 ARI cases and 225 deaths (in 2017)

(CBHI, 2018). This increase in deaths due to ARI were directly linked to the increased PM concentration in this region making the state victimized for major number of deaths among all the northeastern states.

A strong urban and rural division in this region might result in significantly varied location-specific dominating sources. Better understanding of the sources, can help in developing better mitigation strategies which is the accelerating need of the hour in this region. However, in a region like Brahmaputra valley of Assam which is in proximity to Himalayan glaciers, the seasonal variations of both size resolved aerosol dry and wet deposition of PM has not yet been reported. Very few studies have been conducted on size segregated aerosols focusing only urban areas thus raising the demand for the expansion of studies towards rural areas also. Moreover, there is a great requisite for characterization and source apportionment of the size resolved PM, which is highly neglected in most of the studies. Therefore, a long-term measurement and the characterization of size resolved aerosol dry and wet deposition in this region would gain access in understanding the sources, seasonal characteristics and impacts of urban developments in the valley.

1.3 BROAD OBJECTIVES OF THE STUDY

Main objective of the study is to effectively investigate the dominant sources that are responsible for the air quality deterioration in this region. For this purpose, wet deposition in an urban area and dry deposition in both urban and rural areas of Assam were conducted. Independent laboratory testing techniques were employed to comprehensively characterize the samples collected for both wet and dry PM. Also, the regional deposition of size resolved PM mass in human respiratory tract were evaluated using the inhalation and deposition curves. Using the obtained results, receptor-oriented analysis was employed to predict the sources and their contributions for both wet and dry depositions so as to a have the holistic idea of predominant sources in this region which can further aid in the development of air pollution abatement strategies.

1.4 WORK PLAN

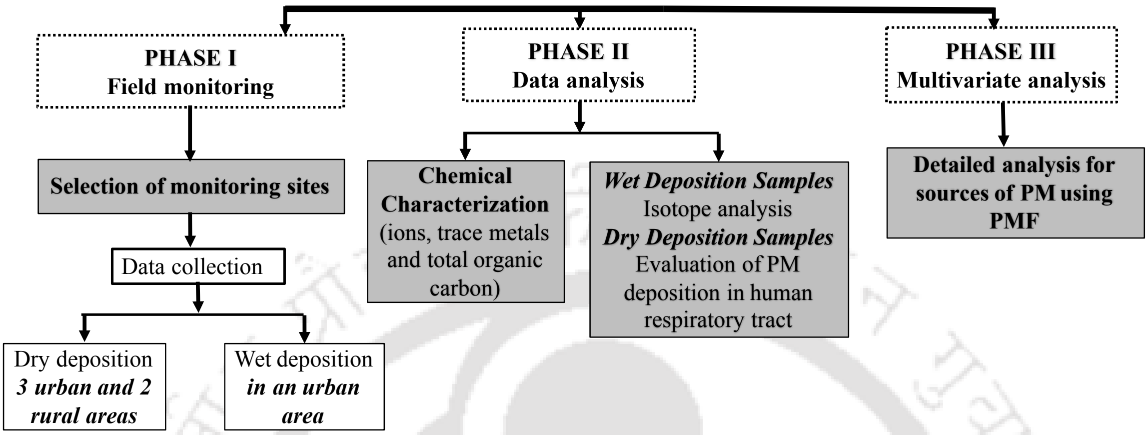

Fig 1.4 Work flow used to estimate dominant sources and their contributions for both wet and dry depositions of PM in Assam

1.5 NOVELTY OF THE STUDY

Being one of the richest biodiversity hotspots and center for two UNESCO world heritage sites, the state of Assam is experiencing the highest mortality rate among all the northeastern states of India, due to rising PM concentration (CBHI, 2018; Chatterjee et al., 2006). Moreover, the variations in climatic and geographical conditions of northeast India can have large scale impacts. Such an atmospheric crisis demands a serious concern in executing methodologies to circumvent the air quality issues in this region. Thus, special emphasis is to be given in neutralizing the adverse effect of PM in this region. Therefore, the current study assesses the impact of dry and wet deposition of PM, its major components, important sources, focusing both urban and rural areas. This kind of constructive approach not only gives a holistic idea of total atmospheric deposition in this locality but also suggests the various promising aspects for the betterment of air quality and execution of better mitigation strategies.

1.6 ORGANIZATION OF THESIS

The present thesis is divided in to five chapters. After the introduction to the subject in the first chapter, further chapters are divided as follows:

Chapter 2 provides a detailed review of literature pertinent to the source apportionment studies of wet and dry depositions of PM carried out in various parts of the India. The chapter concludes with a critical appraisal of literature and a detailed scope of the work.

Chapter 3 describes in detail the monitoring sites selected for the study, method of sampling and the experimental approaches adopted.

Chapter 4 presents the detailed analysis of wet deposition samples and the compilation of the results obtained. The chapter also includes the relevant analysis, results and discussions of size resolved PM collected in this study.

Chapter 5 summarizes the critical findings obtained from the study, limitations of the work and identifies the areas for future study.

CHAPTER II – LITERATURE REVIEW

2.0 GENERAL

Aerosols, being chemically heterogeneous with a mixture of different constituents, are often treated as terminating sites for many gas phase chemical reactions. It constitutes different cations (SO_4^{2-}, NO_3^-, Cl^-, PO_4^{3-}, F^-), anions (Na^+, Mg^{2+}, Ca^{2+}, K^+, NH_4^+), trace metals (Al, Cd, Co, Cr, Cu, Fe, Ni, Mn, Pb, Sr, Zn), and organic carbon and elemental carbon (OC and EC) (Lai et al., 2017). One of the chief reasons for the health effects associated with PM is the production of free radicals by the toxic metals which affect the Deoxyribonucleic acid (DNA) and the pulmonary system (Stanek et al., 2011; Valavanidis et al., 2008; Xiaoyan et al., 2015). For example, Cd at short term exposure causes pulmonary and bronchial irritation while the long term exposure can damage liver, bones, blood and the nervous system (Geiger and Cooper, 2010; Martelli et al., 2006). Cu is an essential nutrient but when uptaken in excess amounts causes liver damage (de Romaña et al., 2011). Cr (VI) is carcinogenic and affects blood element homeostasis, altering Fe metabolism and causes renal injury (Song et al., 2012). Exposure to longer duration of the neurotoxic element, Mn, results in a neurological disease called manganism (Aldape et al., 1999). Ni causes dermatitis and respiratory disorders (Lu et al., 2009). Pb, even at lower concentrations affects the nervous and vascular systems (Vahter et al., 2007). Zn is an essential nutrient but constant exposure leads to anemia (Geiger and Cooper, 2010). In addition to health effects, some of these components play a prominent role in radiative forcing (Pilinis et al., 1995).

Few present-day sampling networks in developed countries, like Speciation Trends Network (STN), the Interagency Monitoring of Protected Visual Environment

(IMPROVE) and the Southeastern Aerosol Research and Characterization (SEARCH) has contributed to a greater extent that they output the real time speciated aerosol data (Reff et al., 2007). Unlike these developed countries, where chemical speciation networks consistently monitor the various components of PM at different locations, only total masses of PM_{10} and $PM_{2.5}$ are measured in India (Gargava and Rajagopalan, 2016). Moreover, to design effective control strategies for PM in a region, it is imperative to understand its temporal variations, composition and sources.

Source apportionment technique is the method which involves the identification of pollution sources and quantification of their contributions based on source and receptor characteristics. For this purpose, three major approaches are employed: chemical transport model, receptor oriented analysis and emission inventories and dispersion modelling (Belis et al., 2013). Among these, receptor models are most commonly used around the world for identification of sources and their contributions (Querol et al., 2001; Schauer et al., 1996; Viana et al., 2008; Zheng et al., 2002). These are broadly classified to chemical and microscopic methods, of which the usage of latter method, limits in large-scale measurements and confines in identifying inorganic compounds (Shi et al., 2008). Amongst the many chemical models, the chemical mass balance (CMB) model is used frequently, although it has limitations such as the unavailability of locally determined source profiles (Watson et al., 2001). The receptor models developed using the factor analysis approach (Table 2.1), i.e. principal component analysis (PCA), Enrichment factor (EF) positive matrix factorization (PMF) and Unmix (UNMIX), solve the problem with CMB. In the last decade, different Indian studies used EF (34%), PCA (36%), PMF (15%), UNMIX (3%) and CMB (10%) to resolve the dominant sources and their contributions of PM (Singh et al., 2017).

Table 2.1 Principles and limitations of different approaches of receptor models (after (Banerjee et al., 2015))

Method	Principle	Limitation
CMB	• Complete source profile information processed through multiple linear least squares approach • Choices of source profiles should avoid collinearity	• Does not apportion secondary particulates • Assumes every single particulates source have been identified • Non-reactive, stable and uniform tracer
PCA	• Convert observations to a set of linearity uncorrelated variables • Principal component with maximum variance are interpreted as most influential source	• Principally based on statistical association of data rather than chemical nature • Does not perform data uncertainty treatment
PMF	• Consider known experimental uncertainties as input to resolve weighted factorization using non-negativity constraint • Does not essentially require source profile information	• Require large particulate dataset • Sensitive to model pre-set assumptions • Factor source relationships needs to be improved
UNMIX	• Applies singular value decomposition to find out edges useful to reduce data dimension • Works on edges detection technique to generate source contribution by geometrically driven approach	• Does not count on measurement uncertainties or process samples with missing data sets • Require large particulate dataset
ME	• Multilinear problem is represented by set of equations approximating single data representing different unknowns • Includes heterogeneous particulate information and uncertainties with modifications flexibilities	• Typically assume linear relationships between all variables • Sensitive to model preset parameters • Require large particulate dataset
EF	• Compares the relative ratio of elemental composition in the measures sample to corresponding ratio in the natural background composition	• Better useful for source identification rather than quantification • Unique and strong tracer information are required

Identification of key tracer species for each source is one of chief steps in source allocation. There is ambiguity, as reported by earlier reviews in finding the tracers (Banerjee et al., 2015; Karagulian et al., 2015; Pant et al., 2016; Singh et al., 2017). So, based on the commonly reported sources in different studies, seven common sources categories along with the commonly used tracers, were considered: vehicular emissions, biomass burning, coal combustion, industrial emissions, marine source, dust emissions and other sources.

(i) Vehicular emissions: This includes on-road emissions related to motor vehicles, i.e. tail pipe, non-tail pipe, brake wear and tire wear (Cu, Pb, Zn, Ni, Fe, Co, TOC).

(ii) Biomass burning: Emissions from burning of biomass, wood, vegetative burning, etc., which are used for cooking and heating (K^+, SO_4^{2-}, NO_3^-, OC).

(iii) Coal combustion: Emissions from mining activities and burning of coal (Cl^-, F^-, SO_4^{2-}, OC, Pb, Sr, Zn, Cd).

(iv) Industrial emissions: This group contains the mixture of emissions mainly from power plants, and all the various types of industries (Cu, Co, Cr, Cd, An, Mn, Cr, Fe, Ni, Pb).

(v) Marine source: Sea salt and ship emissions (Cl^-, Na^+, Mg^{2+}, K^+).

(vi) Dust emissions: This category includes emissions from re-suspended dust, earth's crust and other soil emissions (Ca^{2+}, Mg^{2+}, Fe, K^+, Al, Mn, Na).

(vii) Oil refinery: This includes the emissions from oil refinery plants that refines the crude oil in to petroleum products (Ni, Cu).

(viii) Other sources: All other anthropogenic emissions, including contributions of secondary organic aerosols (SO_4^{2-}, NO_3^-, NH_4^+).

Therefore, this chapter presents a consolidated view of the literature review based on broad objectives of the study. The chapter is categorized in to two main sections. The first section discusses the wet deposition studies carried out in different locations of India. Second section reviews the studies on dry deposition of PM within India. The chapter concludes with a summary of critical appraisal of the literature and the objectives formulated based on the literature survey.

2.1 STUDIES ON WET DEPOSITION

Time trend analysis in precipitation (rainwater) chemistry has gained importance due to its potential in environmental monitoring such as, acid deposition (acid rain), trace metal deposition, eutrophication as well as analysis of the global climate change (Xiao, 2016). Though this practice has been extensively used in developed countries, however, only a few workers have explored its potential in developing countries like India (Kulshrestha et al., 2014; Tiwari et al., 2012).

Innumerable studies on wet scavenging of atmospheric pollutants have been reported in India targeting the metropolitan cities, including northern India especially in the nation's capital, Delhi (Rao et al., 2016), in western India (Budhavant et al., 2014; Momin et al., 2005), in eastern India (Das et al., 2005; Kulshrestha et al., 2014) and southern India (Kulshrestha et al., 2003). Also, there exists continuous monitoring programs such as Background Air Pollution Monitoring Network (BAPMoN), Global Atmospheric Watch (GAW) and Precipitation Chemistry Monitoring Program of the Indian Institute of Tropical Meteorology (IITM) in Pune. However, main focus of these studies was the rainwater characterization instead of source identification. The rainwater composition is an important attribute that helps us to quantify the relative contributions of different sources of atmospheric pollutants. The rainwater composition changes from region to region due to the difference in the sources. Therefore, findings from one location cannot be inferred to other locations, without having the monitoring data, due to the variation of transboundary characteristics of the atmospheric pollution (Akpan et al., 2018). Previous studies reported that rainwater consists of mixture of chemical species originated from natural (sea salt (Na^+, Cl^-) and soil dust (Ca^{2+}, K^+, Mg^{2+})) or anthropogenic sources (industrial and vehicular emissions) emits acidic species such as SO_4^{2-} and NO_3^-, which are carried away by wind from distant sources (Hamilto-Taylor and Willis, 1990; Jonnalagadda et al., 1994).

The variation in the chemical composition of rainwater depends on sources, long range transport of air masses, meteorological conditions (Herrera et al., 2009; Kulshrestha et al., 1999; Zunckel et al., 2003). Multivariate receptor techniques are used to identify the sources and their contributions of rainwater (Qiao et al., 2018; Qiao et al., 2015a; Qiao et al., 2015b). In Varanasi, dominant sources namely industrial emissions, crustal sources and biomass burning were extracted by means of principal component analysis (PCA) (Pandey and Singh, 2012). Using PCA, three significant sources, soil dust, sea salt and fossil fuel combustion were reported as major sources, affecting rainwater chemistry, in Nainital, in the central Himalayas, India (Bisht et al., 2017). This was widely used model in many of the South Asian urban cities (Chakraborty et al., 2016a; Rao et al., 2016; Roy et al., 2016; Tiwari et al., 2016). Rao et al. (2016) used the US EPA's PMF model and reported that the rainwater composition was greatly influenced by soil dust (with Ca^{2+} as dominant cation) and fossil fuel consumption (SO_4^{2-} as predominant anion).

In contrary to only few previous studies (For example, see (Balachandran and Khillare, 2001; Kulshrestha et al., 2014)) witnessing acid rain events, most of the studies (For example see (Al-Momani et al., 1995)) reported the alkaline nature (pH>5.6) of rainwater due to its neutralization by airborne dust and ammonia released from natural and anthropogenic sources. However, most of these studies were conducted in monsoon periods (Budhavant et al., 2011; Khemani et al., 1994; Momin et al., 2005) but little attention was paid to non-monsoon rains (Rao et al., 2016). Missing the analysis on non-monsoon rains could result in erroneous conclusions on rainwater chemistry (Kumar et al., 2014b).

2.2 STUDIES ON DRY DEPOSITION

Metadata reported by the source apportionment studies carried out throughout India on individual masses of PM, along with special emphasis on episodic analysis and size resolved PM are presented briefly in this section.

2.2.1 Diverse sources of PM over India

Fig 2.1 shows the source contributions to PM in different cities over India. PM concentrations in northern and eastern India were higher than cities in other parts of the country. These studies are further divided into residential, city centre and industrial areas in different regions of the country as described below.

Northern India

Around 19 source apportionment studies were carried out at Delhi in last decade with 37% in the last five years. Source apportionment studies in other cities were fewer in number.

Delhi

Delhi, the capital city of India, often features in the world's top polluted cities list. Around 30% of its population was diagnosed with respiratory disorders caused due to air pollution (Kandlikar and Ramachandran, 2000) which led to 18,600 premature deaths every year (TERI, 2001).

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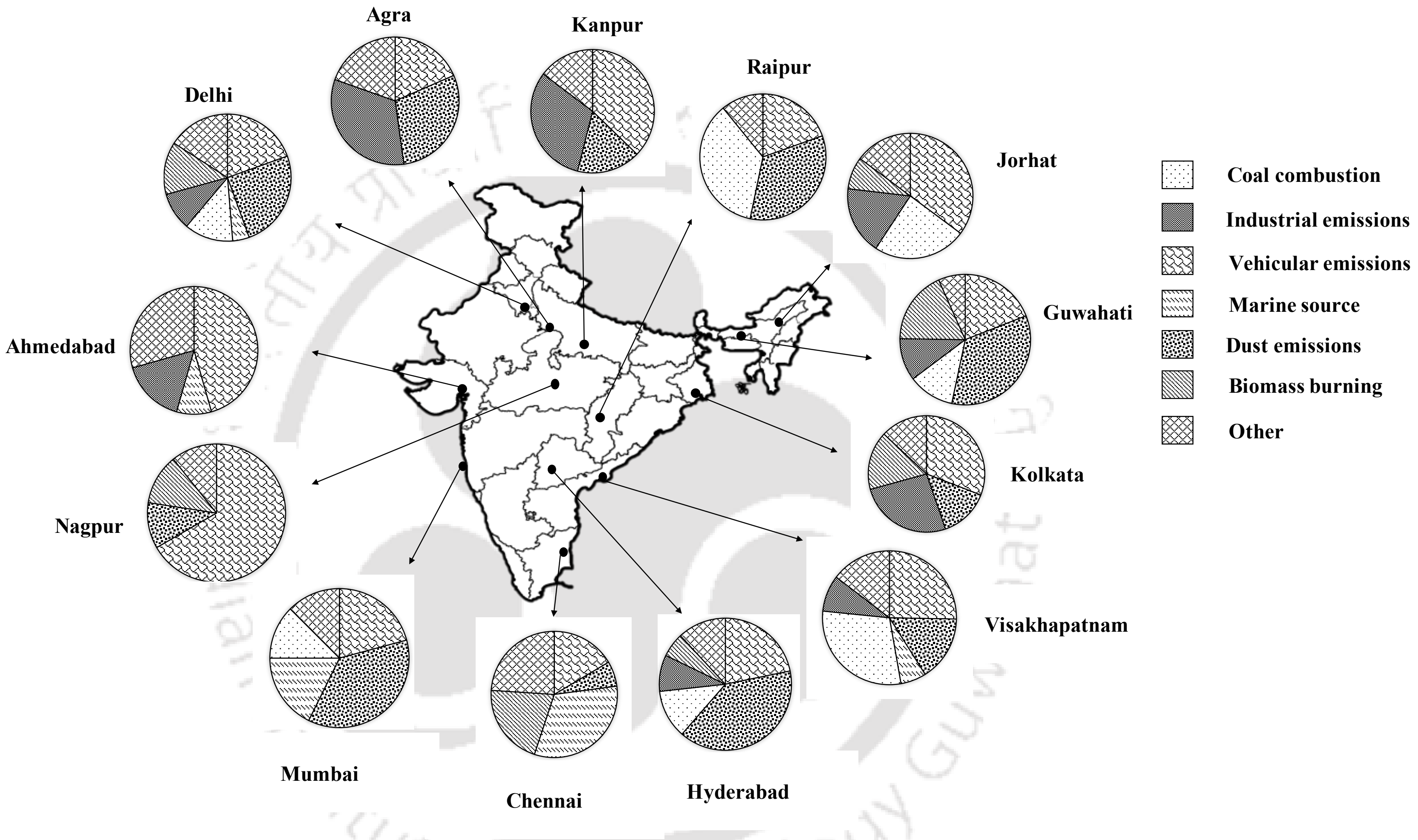

Fig 2.1 Relative source contributions (%) of PM in major cities in India.

Residential area

In Delhi most commonly found sources were vehicular emissions (23%), industrial emissions (29%), biomass burning (15.02%) and dust emissions (35%) (28, 29, 31, 32).

City centre

Vehicular emissions (20.3%), biomass burning (13.95%), industrial emissions (10.04%), dust emissions (25.89%), coal combustion (12.94%) and marine source (4.4%) were the major sources near city centers in Delhi (28, 29, 33, 34, 35).

Industrial area

Most important sources were industrial emissions (46.2%), dust emissions (22.8%) and vehicular emissions (10.2%) (28).

Agra

Residential area

Source apportionment studies conducted in residential areas of Agra (Habil et al., 2013; Kulshrestha et al., 2009), one of the most polluted cities in Uttar Pradesh, identified vehicular emissions (28%) and dust emissions (48%) as the major sources contributing to PM_{10}. While to $PM_{2.5}$, industrial emissions (30%), vehicular emissions (17%), dust emissions (27%) and other anthropogenic activities (18%) were the main sources. Dominant sources to water soluble ions in total suspended particles (TSP) were biomass burning and local soil (Satsangi et al., 2013). Apportionment of PM sizes and trace metals in this region found the following sources (Habil et al., 2013): vehicular emissions and soil dust (38.1%), fossil fuel combustion (25.8%), garbage burning and other activities (25.7%).

City centre

Source apportionment study of PM_{10}, identified that vehicular emissions (27.45%), biomass burning (13.67%), industrial emissions (13.52%) and dust emissions (30%) as the dominant sources contributing to PM_{10} (36, 37). Concentrations of PM sizes and trace metals identified the following sources (Habil et al., 2013): vehicular emissions and soil dust (47.6%), vehicular wear and tear (33.8%) and biomass burning (17.9%).

Kanpur

Kanpur, which is located in central Indo-Gangetic plain (IGP) region, poses constant threat of varying emission sources thereby leading to higher concentrations of PM. One of the studies conducted in a residential area, (Chakraborty and Gupta, 2009) in this region found out vehicular emissions (38.78%), industries (32.52%) and dust emissions (17.07%) as major sources of PM_1. Another study in the same location, found that anthropogenic sources were crucial during winter, while crustal emissions are high in summer (Ram et al., 2012).

Eastern India

Kolkata

Population in Kolkata, one of the megacities in India, is suffering from 41.3% and 47.8% of upper and lower respiratory issues, respectively (WBPCB, 2012). Number of source apportionment studies carried out in Kolkata are 57% lesser than Delhi.

City centre

In order to identify sources of total suspended PM, road side locations in Kolkata were studied and found that vehicular emissions (42%), dust emissions (17%) and industrial emissions (7%) were the major sources (Kar et al., 2010). Extensive sampling conducted in this megacity to understand the sources of $PM_{2.5}$ using PCA, identified vehicular emissions (38%), biomass burning (27%), dust emissions (18%) and other secondary anthropogenic (11%) as the dominant sources (Chatterjee et al., 2012).

Residential and Industrial

One of the studies conducted in 16 different locations of Kolkata (i.e. residential areas close to traffic junctions, coal fired power plants, industrial belts, waste incineration plants, cement factories and brick kilns) identified major sources as vehicular emissions, industries and coal combustion using PCA (Das et al., 2015).

Source apportionment studies in northeast India were mainly concentrated in the mid Brahmaputra valley region. One of the studies in suburban area, using enrichment factor (EF), found out that vehicular emissions (38%), coal combustion (26%) and industrial emissions (19%), biomass burning (9%) were the dominant sources (Khare and Baruah, 2010).

A study at a residential region (Deka et al., 2016), using PCA-multiple linear regression (MLR), marked three major sources: biomass burning (23%), dust emissions (26%) and vehicular emissions (22%).

Western India

Mumbai

Mumbai, the industrial capital of India, which is growing in all the commercial activities leading to the deterioration of air quality day-by-day. To understand these increasing sources, several source apportionment studies have been conducted in the past which are well discussed in the following section.

Residential area

Sources to PM_{10} were dust emissions (35%), seasalt (17%), coal combustion (12%) and vehicular emissions (20%) and for $PM_{2.5}$ were dust emissions (26%), industrial emissions (4%), seasalt (14%) and coal combustion (7%) (Kothai et al., 2011).

City centre

In this city, major sources observed were dust emissions (11%) and marine source (11%) for $PM_{2.5}$, and for PM_{10}, vehicular emissions (22%), biomass burning (18%), marine source (19%) and dust emissions (25%) (Kothai et al., 2011).

Industrial area

In this region, dominant sources found were dust emissions (10%) and marine source (8%) for $PM_{2.5}$, and for PM_{10}, vehicular emissions (25%), biomass burning (15%) and dust emissions (27%) (Kothai et al., 2011).

Ahmedabad

Ahmedabad, a semi-arid and well populated area in western India, has all types of settings like residential, small and large scale industries, commercial activities etc. Identification of the major sources to TSP using PMF revealed that dust emissions (57%), biomass burning (10%), vehicular emissions (17%) and marine source (5%) are the resolved factors (Raman et al., 2010). Study using PMF, in an city centre reported the dominant sources of PM_{10} as dust emissions (37%) and biomass burning (33%), and industrial emissions (11%), marine source (6%) and vehicular emissions (31%) for $PM_{2.5}$ (Sudheer and Rengarajan, 2012).

Nagpur

Nagpur, the centrally located fast growing metropolis city of India, is also victimized of poor air quality. A recent source apportionment study in which $PM_{2.5}$ sampling was carried out in Nagpur using CMB found the following major sources: vehicular emissions (57%, 62% and 65%), biomass burning (15%, 11% and 9%) and dust emissions (6%, 10% and 7%) in residential, commercial and industrial regions, respectively (Pipalatkar et al., 2014).

Southern India

Source apportionment studies over South India have immensely increased during the last decade and is extensively concentrated in Chennai, Hyderabad and Visakhapatnam.

Chennai

Sources to PM in a city centre was studied and identified as dust emissions (74%), marine source (16%) and vehicular emissions (10%) (Srimuruganandam and Nagendra, 2011). Another study conducted in an urban site using PMF reported that marine source (40.4% in PM_{10} and 21.5% in $PM_{2.5}$), vehicular emissions (20% in PM_{10} and 11% in $PM_{2.5}$), biomass burning (0.7% in PM_{10} and 14% in $PM_{2.5}$), and dust emissions (3.4% in PM_{10} and 4.3% in $PM_{2.5}$) are the major sources (Srimuruganandam and Shiva Nagendra, 2012).

Hyderabad

Hyderabad, an emerging metropolitan 400-year-old city, has an increasing trend of urbanization since 1960 due to which a significant degree of air quality decline is being witnessed. Source apportionment study in the city centre identified dust emissions (40%), vehicular emissions (22%), coal combustion (12%), industrial emissions (9%) and biomass burning (7%) as dominant sources for PM_{10} and vehicular emissions (31%), dust emissions (26%), coal combustion (9%), industrial emissions (7%) and biomass burning (6%) as the source contributors to $PM_{2.5}$ (Gummeneni et al., 2011). In a residential area, the common sources found were dust emissions (36% and 20%), vehicular emissions (41% and 38%), biomass burning (6% and 9%) and coal combustion (6% and 12%) for both PM_{10} and $PM_{2.5}$, respectively.

Visakhapatnam

Visakhapatnam, also known as the financial capital of state of Andhra Pradesh, had gradual decline of air quality and thereby turning out to be air pollution hotspot. Source apportionment of PM_{10} showed dust emissions (22.5%), seasalt (9.7%), coal combustion (15.5%), industrial emissions (5.1%) and biomass burning (35%) in residential sites, and dust emissions (22.5%), seasalt (5.5%), coal combustion (26.1%), industrial emissions (7.8%) and vehicular (14%) in industrial sites (Police et al., 2016).

Central India

Durg

Source apportionment studies in other cities of the Central India are few. At a city centre of Durg in Chhattisgarh, source identification study (Deshmukh et al., 2011) using PCA revealed two principal components which explain 76.6% and 65.9% of the variance for $PM_{2.5}$ and PM_1 respectively. One component had coal combustion, vehicular emissions and biomass burning (52% and 45%) and another was dust emissions (25% and 21%).

Raipur

Study near city centre, using PCA (Deshmukh et al., 2013b), revealed that while coal burning (33.2%), dust emissions (31.6%) and vehicular emissions (18.4%) were prominent sources contributing to $PM_{2.5}$, coal burning (50.7%), dust emissions (25.3%) and vehicular emissions (18%) were sources to PM_{10}.

Summary of sources in different regions of India

In residential areas in north, south, east and west the dust emissions (42%), vehicular (41%), vehicular emissions (30%) and dust emissions (43%) were the dominant sources. While in city centres in north, south, east, west and central India, dust emissions (28%), dust emissions (47%), vehicular emissions (40%), vehicular emissions (42%) and coal combustion (45%) were the main sources. In industrial regions in north, south and west, industrial emissions (46%), coal combustion (26%) and vehicular emissions (44%) were important sources, respectively. Overall in India dust emissions (30%) and vehicular emissions (31%) are the main contributors to PM.

2.2.2 Diverse sources of PM during episodic analysis over India

Concentrations of regulated air pollutants exceed Indian National Ambient Air Quality Standards (NAAQS) in most of the cities in India (Kota et al., 2018). Furthermore, episodes such as burning of fireworks which lead to the release of large amounts of harmful gases and toxic substances in to the atmosphere thereby contaminating the air and bringing adverse effects to the human health worsen the air pollution. Rapid increase in the pollutant concentrations due to fireworks have been previously reported (Gouder and Montefort, 2014). Determination of PM_{10} (particulate matter with aerodynamic diameter < 10 µm) in the ambient air is essential as pollution due to fireworks causes pulmonary effects (Verma and Deshmukh, 2014), and its impact is acknowledged mostly in the case of young children, elderly persons and pregnant women (Makri and Stilianakis, 2008). Fireworks are made up of various organic and inorganic chemicals thereby creating smoke plumes that contain charcoal, sulphur, potassium, lead, aluminum, iron and barium nitrate (Kulshrestha et al., 2004; Steinhauser et al., 2008). Chemical reactions propel and burst them into special shapes. While, Na and K are used as metal oxidizers, Zn is used to produce smoke effects and Sr is used to stabilize fireworks (Kulshrestha et al., 2004). Pollutants released from fireworks at a higher altitude are diluted before coming into contact with human populations, which can reduce health impacts (Betha and Balasubramanian, 2013).

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However, the ground level fireworks display have an immediate impact on human health. Acute eosinophilic pneumonia (AEP) has been tested positive for a patient due to inhalation of smoke continuously for three nights from burning of fireworks (Hirai et al., 2000). While fever, cough and dyspnea were reported initially as an acute effects (Sharma et al., 2015), pollution due to fireworks causes chronic respiratory and cardiovascular diseases, pulmonary effects, premature death and cancer (Barman et al., 2008; Nasir and Brahmaiah, 2015). Patients with wheezing, respiratory diseases, exacerbation of the bronchial asthma and bronchitis increased by 30% to 40% during the Diwali celebration in India (Gouder and Montefort, 2014).

Several studies have been carried out in different parts of India to assess the impact of the fireworks burning on PM_{10} related air quality. A short term study reported 2-3 times elevation in PM_{10} concentrations during Diwali festival in Hisar city in north-west part of India (Ravindra et al., 2003). PM_{10} during Diwali day in Lucknow in northern India was reported as 7.53 times higher than normal days (Barman et al., 2008). Five times increase in PM_{10} concentration was observed in Kolkata in eastern India during Diwali (Chatterjee et al., 2013). An uncommon increment in PM_{10} concentration to 35 times during Diwali day in comparison to normal day, due to abrupt pollution load caused by burning of fireworks was reported in Vadodara in western India (Nasir and Brahmaiah, 2015). In Rajnandgaon in central India, PM_{10} concentration during Diwali period has raised to nearly 3 times than normal days (Ambade and Ghosh, 2013). 4 to 10 times increase in PM_{10} concentrations were observed during Diwali in Nagpur in central India (Khaparde et al., 2012). Owing to already high PM_{10} concentrations, several studies were conducted in the national capital, New Delhi. For example, high concentrations of PM_{10} i.e. 767 $\mu g/m^3$ and 620 $\mu g/m^3$ were observed during the Diwali festivals of 2008 and 2009, respectively (Perrino et al., 2011). Also a community based health survey (Sharma et al., 2015) conducted during Diwali in

2013 revealed an increase in number of patients in Delhi with problems related to respiratory diseases, hearing issues, irritation in eyes and headache.

A previous study in northeast India observed increase in concentrations of metals, anions and cations during festive days compared to other days (Deka and Hoque, 2014). In Raipur, the concentrations of ions were 10 times greater than the normal days (Pervez et al., 2016). Similar increase in the metals associated with fireworks burning were reported in Delhi (Sarkar et al., 2010).

Only two investigations till date were reported from northeastern India. In Tezpur, mean PM_{10} concentration observed was 87.45 $\mu g/m^3$ which was 2.13 times more than the normal day concentrations (Deka and Hoque, 2014). Another study from Dibrugarh (Pathak et al., 2015), reported that PM_{10} and $PM_{2.5}$ concentrations during Diwali were 168 and 160 $\mu g/m^3$ respectively which was 5.33 and 5.74 times higher than the normal day concentrations. However, no health correlation and risk level study was carried out in those studies. Over the years, these celebrations during Diwali have only gained more importance. However, studies in a residential setting with a goal of understanding direct environmental impact of fireworks on health of residents are rare.

2.2.3 Studies on size resolved PM in India

In India, during last few decades, intensive research had been carried out on the independent size fraction of PM either 2.5 or 10 µm, gaining knowledge on their composition, seasonal variation of their mass concentrations and source contributions (Singh et al., 2017). Many reported that the coarse PM is mainly due to resuspended soil fraction and is considered as major problem in local or urban scale (Banerjee et al., 2015). Whereas, $PM_{2.5}$ and PM_1, having longer residence times originates from different anthropogenic emission sources, mainly contributing to urban air pollution in India, are viewed as regional or transboundary issue (Shin et al., 2009). However, the findings reported by many Indian studies highlighted the notable importance of investigating the size resolved PM, to understand the transformation mechanism of aerosols, which influence the changes in their concentrations and compositions as they are released into the atmosphere, so as to anticipate the long-term mitigation policies (Kota et al., 2018).

In recent years, few studies Fig 2.2 have been conducted in India focusing mainly on seasonal variation of the size resolved mass concentrations, chemical composition and sources in major cities like Delhi (George et al., 2017; Kumar et al., 2018), Rajim (Nirmalkar et al., 2016), Raipur (Deshmukh et al., 2013a), Durg (Deshmukh et al., 2013c), Nagpur (Pipalatkar et al., 2012) and Kolkata (Nag et al., 2005).

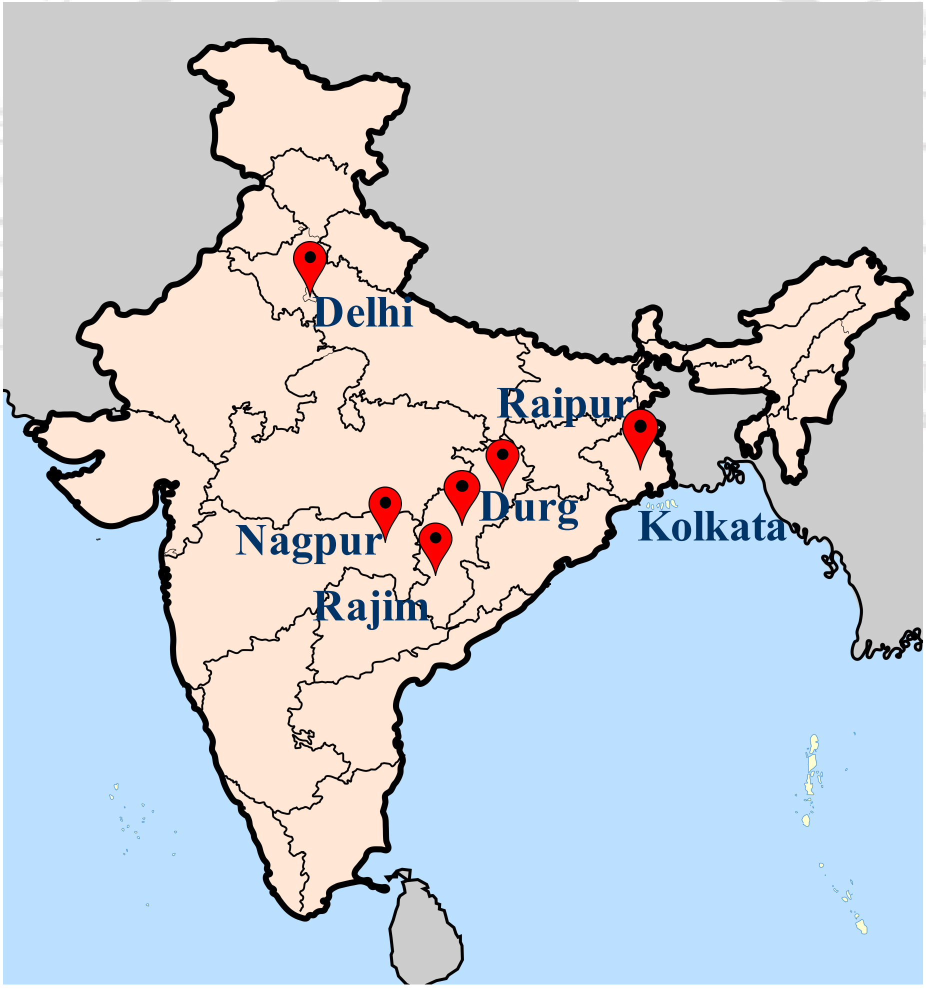


Fig 2.2 Studies conducted on size segregated PM in different parts of India

However, very few studies have focused on the assessment of deposition in human respiratory tract restricting to particle mass with not much information on the deposition of chemical species and sources that are directly linked up to cause of adverse effects on human health (Gupta and Elumalai, 2017; Nag et al., 2005). Most of the size segregated PM studies have been conducted in urban areas with limited or no information in background sites which determines the influence of long-range transport of pollutants on the atmosphere (Chelani et al., 2010; Kumar et al., 2018).

2.3 CRITICAL APPRAISAL OF LITERATURE

- Size, chemical composition and related sources of PM is directly linked to various health effects due to their deposition in human body which thus demands performance of site specific investigations.
- In Indian scenario, most studies focused mainly on seasonal variation of the individual mass concentrations of PM, their chemical composition and sources with few independent studies carried out on size resolved PM. Also, very few studied the assessment of particle deposition in human respiratory tract with no information on deposition of chemical species and sources.
- In a region like Brahmaputra valley of Assam (northeast India), very few studies have been conducted till date to estimate the concentrations of PM, its components and the dominant sources. Seasonal variation of size resolved PM and its composition at a single location in this region was not yet reported.
- Studies were conducted on either wet or dry deposition of PM, in either urban or rural areas. The year-long information of size resolved PM in representative sampling locations and their particle deposition along with source apportionment is not well documented in this region.

- Also, only limited wet deposition studies were conducted in northeast India with missing analysis during non-monsoon period. Similarly, heavy metals and total organic carbon analysis which generally reveals particular source types were rarely carried out.

- All the other Indian cities reported the alkaline nature (pH>5.6) of rainwater due to its neutralization by airborne dust, with acid rain events witnessed only in northeast India.

2.4 OBJECTIVES AND SCOPE OF THE STUDY

Assam being one of the world's UNESCO sites lies in vicinity to Himalayan glaciers. The earlier studies suggested that the state has been witnessing higher concentrations of PM and acid rain events, indicating its region to be at stake. Hinged on the extensive literature survey carried out, it is concluded that the PM concentration is site specific. Thus, for significant reduction of air pollution in a given locality, the reliable estimation of source contribution and their deposition is crucial. Therefore, samples of wet in an urban and dry PM in different urban and rural locations of Assam were collected for which no comprehensive data are available till date. The main objective of the study is to obtain the holistic idea of atmospheric deposition in this area which will aid in progressing air quality studies in the nearby future. Based on the objective of the study, scope of the work is planned in a systematic way as listed below.

1. Collection of wet deposition samples from an urban area over a period of one year.
2. A year-long monitoring of size resolved PM in three urban and two rural sites of Assam for a period of two weeks in each site, covering three seasons i.e. winter, summer and monsoon.
3. Chemical characterizations of both wet and dry samples collected.

4. Isotope analysis of collected wet deposition samples to understand the origin of source of rainwater droplet during monsoon and non-monsoon seasons.
5. Estimation of regional deposition of size resolved PM in human respiratory tract using the inhalation and deposition curves.
6. Receptor oriented analysis using PMF to understand the dominant sources and their contributions liable for human health effects.

2.5 SUMMARY

The chapter summarized the reviewed literature, pertaining to the objectives of the work. The first section detailed the available wet deposition studies along with their chemical characterization in India. Second section presented a review of source apportionment studies on individual masses of PM, episodic analysis and size resolved PM. The third section presented a brief review of size resolved PM studies conducted in India. The critical appraisal of the literature review is presented, followed by the objectives and scope for the present study.

CHAPTER III – METHODOLOGY

3.0 INTRODUCTION

As per the objectives and scope defined, wet deposition in an urban and size resolved aerosol dry deposition in three urban and two rural areas of Assam, were considered for their detailed and comprehensive analysis. The study utilized highly advanced laboratory testing techniques to accomplish the prescribed objectives. Present chapter is divided in to five sections in which the first one deals with the study areas selected for both wet and dry depositions of PM, along with the sampling procedure, while the second section describes the chemical characterization of collected samples, evaluated through laboratory tests and the third section explains the isotope analysis of wet deposition samples. The fourth section discusses the estimation of regional deposition of size resolved PM in human respiratory tract and finally the fifth section deals with source identification using receptor oriented method.

3.1 STUDY AREA AND SAMPLING

3.1.1 Wet deposition

Study was carried out in urban corridor of Assam, Guwahati (26°11′14″N and 91°41′30″E, with an elevation of 50-680 m), one of the quick developing cities in northeast India. Sampling site is an industrial hub, clustered by specific types of businesses such as an export promotion industrial park, a biotechnology park, a skin and health care industry, a pharmaceutical company and a machine industry. Presence of Indian liquid petroleum gas (LPG) bottling plant in the city premises has further aggravated the worsening of air quality. Besides these, construction activities are carried out around the year. The study area is located 20 km from the heart of the city and is surrounded by major highways.

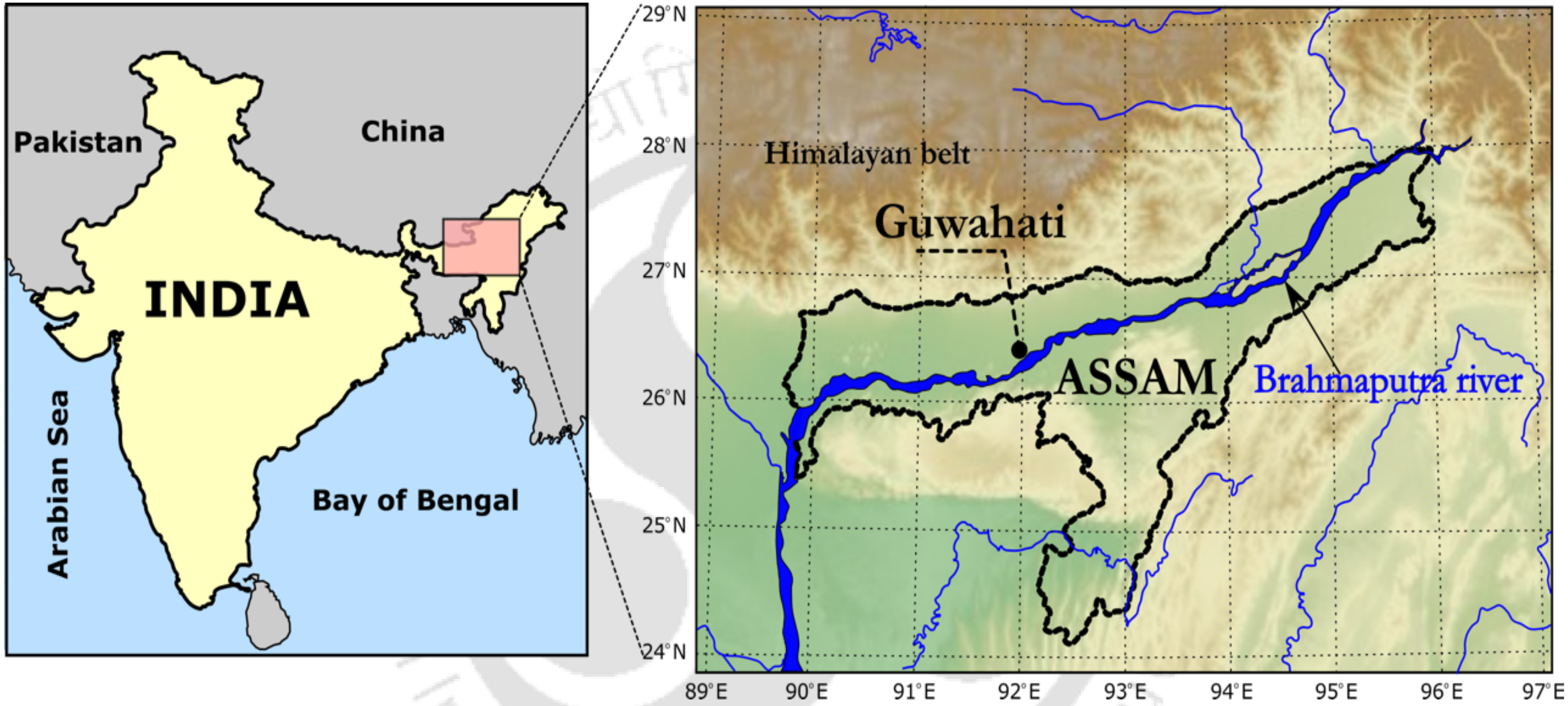


Fig 3.1 Location of the sampling site, Guwahati, the largest city of northeast India

The temperature averages 23.5°C, which peaks in July/August (40°C) and declines in January/December (11°C). The city receives an annual precipitation of 1720 mm, most of which is restricted to the May-October period (http://worldweather.wmo.int/en/city.html?cityId=529). Wet deposition samples were collected on the roof of a building about 10 m from ground level and 50 cm from the floor of the roof. Sampling was done manually using 2 L borosilicate volumetric flask and polyethylene funnel of 20 cm diameter (Khemani et al., 1989; Roy et al., 2016). The collectors (bottles and funnels) were used for sample collection only after proper washing with detergent and HNO_3 followed by deionized water. Collectors were placed as soon as the rain began and were retrieved immediately after the rain stopped. Immediately after retrieval, pH was measured using digital pH meter in an aliquot of the unfiltered sample. Remaining sample amount was transferred into two clean 100 mL polyethylene vials for chemical characterization.

3.1.2 Dry deposition

Total of five locations were considered i.e. three urban and two rural areas of Assam, India. Monitoring sites were selected based on the knowledge of existing air pollutants levels and their pattern within the area. Following are the sites selected for the present study: Jalukbari (S1), Noonmati (S2), ABC Tarun Nagar (S3), Barpeta (S4) and Jorhat (S5). Fig 3.2 depicts the geographical location of these sites and the possible sources in the selected locations. Annexure A describes each selected site along with the prevailing wind direction identified using windrose plot. Also,

Table 3.1 explains the detailed description of the sampling sites selected for the dry deposition of PM along with the activity zones and surrounding activities.

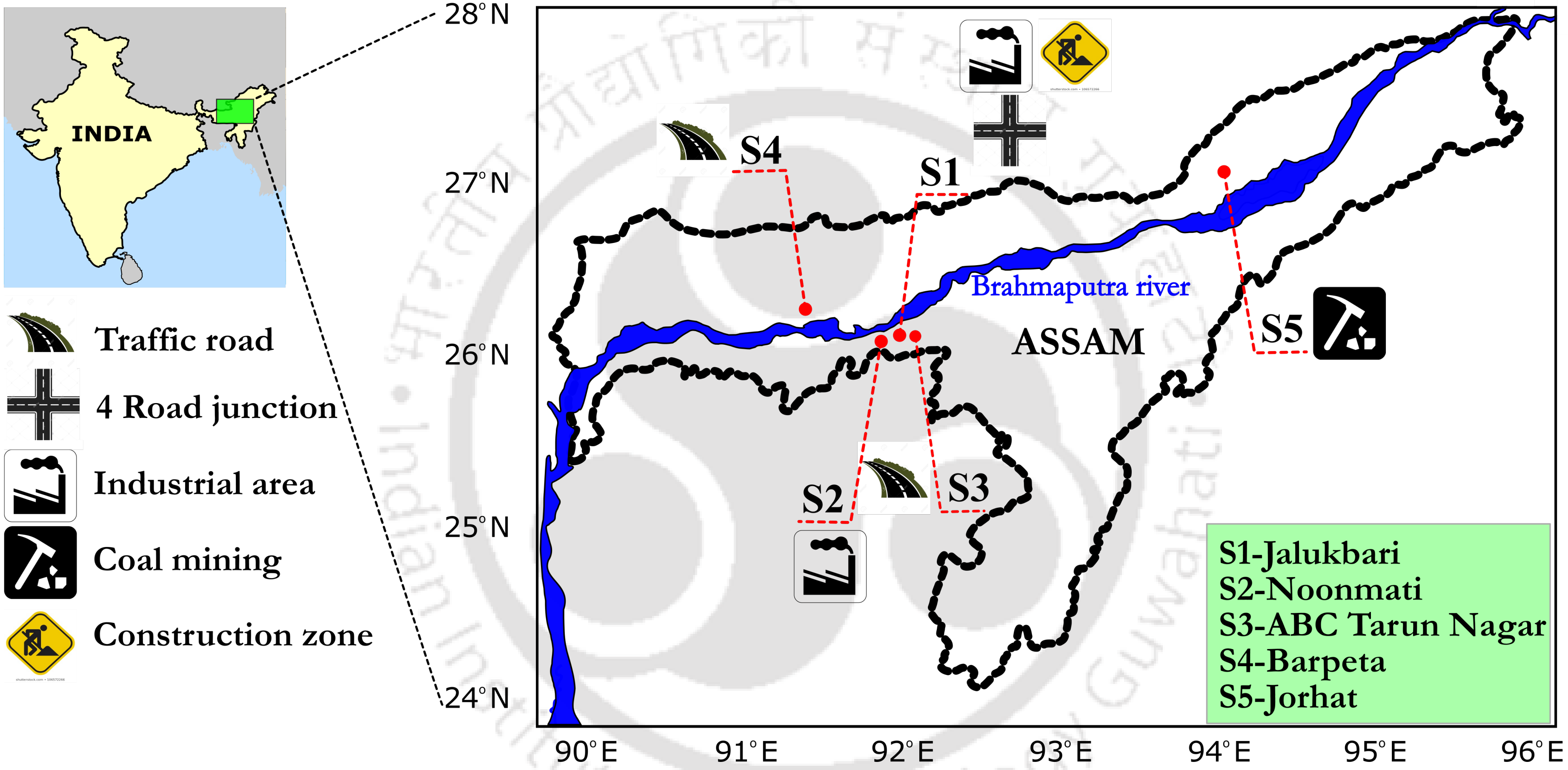


Fig 3.2 Geographical location of all the sites in Assam and possible sources at each site

Table 3.1 Description of the sampling sites selected for dry deposition of PM

Site	Activity zone	Details
Jalukbari (S1)	Residential	Peripheral ring road and well connected by NH 37 having located between Amingaon (south) and Boragaon (north) industrial regions at an aerial distance of 3.5 km and 4 km respectively
Noonmati (S2)	Industrial	Guwahati oil refinery, surrounded by commercial, domestic and vehicular activities
ABC Tarun nagar (S3)	Residential	Traffic congested area mainly connecting the most important city corridor, Guwahati-Shillong road with huge commercial activities and densely populated in the interior parts.
Barpeta (S4)	Rural/background	Mixture of town and village environment, connects to NH 31 with nominal domestic and vehicular activities.
Jorhat (S5)	Mining	Coal mining activities, tea estates, commercial, domestic and vehicular activities.

Sampling was carried out using eight stages non-viable cascade impactor (Fig 3.3) to measure 24 hours size resolved PM concentrations. Whatman® quartz fiber filters, 81 mm diameter baked at 500°C for 4h to remove contaminants, were used to collect the samples. Filters were carried to and fro from the site in sealed polyethylene bags and utmost care was taken to avoid any handling errors. Also, the filter papers were desiccated for 24 hours before and after the sampling to minimize the hygroscopic errors. To prevent the manual contamination of filters, a pair of tweezers were used during their placement and retrieval. The sampling was conducted at a height of 8 m from the ground. Every month, data is being collected at the selected sites, for a period of two weeks. This will aid in having 126 samples at each site in each season and in each location.

Plate 0: >9 µm,
Plate 1: 5.8 – 9 µm,
Plate 2: 4.7 – 5.8 µm,
Plate 3: 3.3 – 4.7 µm,
Plate 4: 2.1 – 3.3 µm,
Plate 5: 1.1 – 2.1 µm,
Plate 6: 0.7 – 1.1 µm,
Plate 7: 0.4 – 0.7 µm,
Filter: <0.4 µm

Fig 3.3 Layout of the 8 Stage Non-Viable Cascade Impactor

(Series 20-800, Thermo fisher Scientific, Inc.)

3.2 CHEMICAL CHARACTERIZATION

3.2.1 Trace elements and total organic carbon (TOC)

Dry deposition: The PM collected on each sample filter was divided into equal aliquots for various purposes. For metals (Al, Cd, Co, Cu, Cr, Fe, Mn, Ni, Pb, Sr and Zn), an aliquot of the sample was digested in a mixture of 5 mL HF, 2.5 mL HNO_3 and 2.5 mL $HClO_4$, on a hot plate under the operating conditions of 100ºC temperature for a duration of 2 hours. Extraction and analysis of metallic elements is carried out as per the U.S. EPA Compendium Methods (U.S. EPA, 1999a; U.S. EPA, 1999b). For further quality assurance, trip blanks were analyzed and necessary corrections were incorporated. Metals recovery test performed using National Institute of Standards and Technology (NIST) standard reference material for 'Urban Particulate Matter-1648a', showed a recovery rate of 87 to 105 % for all the analyzed metals (Annexure B). All dilutions were done using ultrapure water (18.2 grade).

Wet deposition: One part (100 mL) was acidified with 1 mL HNO_3 maintaining pH ~ 2 and used for metal analysis (Co, Cu, Cr, Fe, Mn, Ni, Pb, Sr and Zn). Prior to analysis, each sample was filtrated through membrane filter (pore size: 0.45 µm).

Trace metals were analyzed using Atomic Absorption Spectroscopy (AAS) (Varian Spectra AA-55). Five-point calibration with standards prepared was done for heavy metals analysis using 2% percent HNO_3 solution (Optima Blank-Perkin Elmer Pure Plus) as the reagent blank. $R^2 > 0.99$ was obtained for the calibration curves. Total organic carbon (TOC) was analysed using Shimadzu TOC 5000 total organic carbon analyser equipped with an Automatic Sample Injector (ASI) (Shimadzu, Kyoto, Japan).

Fig 3.4 Chemical characterization of collected wet deposition samples using advanced laboratory testing equipments

3.2.2 Water soluble ions

For this purpose, an aliquot of PM sample was immersed in ultrapure water and ultrasonicated for 20 minutes in case of dry deposition, while in case of rainwater sample, a portion of 25 mL was considered. Samples were stored in prewashed polyethylene bottles and kept at 4ºC until the analysis in Ion Chromatograph (Metrohm 792 basic IC) for the measurement of ions (Ca^{2+}, Mg^{2+}, NH_4^+, Na^+, K^+, F^-, Cl^-, NO_3^-, and SO_4^{2-}). Calibration of the method and quantification of components were carried out using MERCK reference standards (CertiPUR) of 1, 2, 5 ppm for anions and 2, 5, 10 ppm for cations.

Cations were analysed by a cation column (Metrosep C 4 150/4.0). The eluent was prepared in ultra-pure water with 1.7 mmol/L nitric acid and 0.7 mmol/L dipicolinic acid. The eluent flow rate was maintained at 0.9 mL/min. 20 µL of sample was measured using inbuilt loop and injected into the IC system. Before injecting, all the samples were filtered through Millipore 0.22 µm PTFE filters. Calibration and quantification of components were performed using MERCK reference standards (CertiPUR) of 1, 2, 5 ppm for anions and 2, 5, 10 ppm for cations (https://www.merckmillipore.com) (Chaturvedi et al., 2017). All the samples were stored in refrigerator at 4ºC.

To find the acidic neutralizing capacity of the alkaline species (Ca^{2+}, Mg^{2+} and K^+) and check the level of acidification from acid rain pollution in the present study region the neutralization factor (NF) for alkaline species was calculated using Eq 1-3 (Kulshrestha et al., 1995). Therefore, the acid neutralization by Ca^{2+}, Mg^{2+} and K^+ was calculated using their equivalent and non-sea salt (nss) concentrations as follows:

$$NF_{Ca^{2+}} = \text{nss-}Ca^{2+}/(NO_3^- + \text{nss-}SO_4^{2-}) \qquad \textbf{(1)}$$

$$NF_{Mg^{2+}} = \text{nss-}Mg^{2+}/(NO_3^- + \text{nss-}SO_4^{2-}) \qquad \textbf{(2)}$$

$$NF_{K^+} = \text{nss-}K^+/(NO_3^- + \text{nss-}SO_4^{2-}) \qquad \textbf{(3)}$$

The nss values of any particular component (nss-C) were evaluated by using the following equation (Eq 4):

$$[\text{nss-C}]_x = C_x - [Na^+]_x\,[C/Na^+]_{\text{sea salt}} \qquad \textbf{(4)}$$

where, x is sample, C_x is measured concentration of an ion in sample ‘x’, and $[C/Na^+]_{sea\ salt}$ is the seawater ratio for particular ion with Na (Keene et al., 1986).

3.3 ISOTOPE ANALYSIS OF WET DEPOSITION SAMPLES

For rain water samples, isotopic analysis was carried out using 10 ml of the aliquot which was preserved without filtration to avoid any evaporative losses. The isotopic ($\delta^{18}O$ and δD) ratios were determined using a Los Gatos Research (LGR) Liquid Water and Water Vapor Isotope Analyzer (Model: TIWA-45-EP). The isotopic ratios measured, agreed within the ±1σ variability. Further analytical details using the laser isotope technique were presented elsewhere (Chakraborty et al., 2016b). The proportion of stable isotopes is expressed similar to Eq 5 known as global meteoric water line (GMWL) (Craig, 1961):

$$\delta D\ (‰) = 8\ \delta^{18}O + 10\ (R^2 > 0.95) \qquad \textbf{(5)}$$

The slope and intercept of any local meteoric water line (LMWL), which is derived from precipitation collected in any specific area under study, will likely be different from GMWL reflecting the existence of unique moisture source in that region (Craig, 1961). The deuterium excess (d-excess) defined in (Dansgaard, 1964) and expressed as in Eq 6 characterizes the atmospheric precipitation and is mainly useful for source identification.

$$\text{d-excess}\ (‰) = \delta D - 8\ \delta^{18}O \qquad \textbf{(6)}$$

3.4 ESTIMATION OF REGIONAL DEPOSITION OF SIZE RESOLVED PM IN THE HUMAN RESPIRATORY TRACT

The deposition of chemical constituents of PM in ambient particles and human respiratory tract varies significantly and is poorly documented. Fractional particle deposition in different regions of the human body i.e. nasopharyngeal (NOPL), tracheobronchial (TB), and pulmonary (P) is estimated using inhalation and deposition curves (Fig 3.5) assuming normal respiration i.e. tidal volumes of 1450 cm^3 and 15 breaths per minute (Phalen et al., 1991). Deposition fractions for each size range are calculated by integrating the specific deposition curve with an assumption of uniform distribution of particle mass in each size range and the calculation was given in Annexure C.

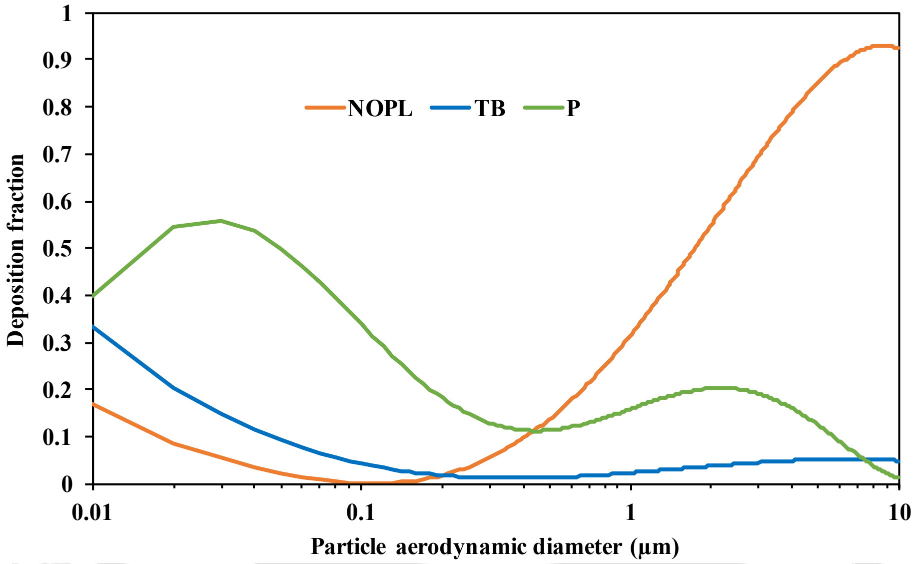


Fig 3.5 Fractional deposition of PM in human respiratory tract

3.5 SOURCE IDENTIFICATION

3.5.1 EF analysis

The EF of trace metals with respect to crustal elements to identify whether certain element is influenced by anthropogenic sources other than its natural sources (Acciai et al., 2017; Rahn, 1971). This technique is applied by taking into account of commonly used reference elements such as Al, Si, Na, Ca, Mg, Mn, K, Fe etc. (Dubey et al., 2012; Yongming et al., 2006). The value of EF is generally calculated as follows (Eq 7):

$$EF_x = [X/Mn]_{Sample} / [X/Mn]_{Crust} \qquad \textbf{(7)}$$

where, $[X/Mn]_{Sample}$ and $[X/Mn]_{Crust}$ are the mean concentration of target element to Mn in the respective sample and in continental crust, respectively. The concentrations of reference element i.e. Mn is based on earth crust mean abundance of the metals taken from CRC handbook (Lide, 2007). The enrichment categories based on EF values are: EF ≈ 1 suggests natural origin, EF = 2-5 is moderately enriched and EF = 5-20 is significant enrichment (Yongming et al., 2006).

3.5.2 PMF analysis

The US EPA's PMF v5.0 model was employed to identify the sources contributing to the pollutant concentrations. Its non-requirement of prior knowledge of contributing source characteristics (i.e. source profiles) improves its feasibility. Based on the given concentration and uncertainty data, PMF provides source profiles and source contributions to each sample and the detailed description is given in the EPA PMF 5.0 guide (Norris et al., 2014). PMF uses the speciated concentration (X_{ij}) of sample '*i*' and species '*j*' and estimates the number of sources '*l*', the source profile '*f*' and the amount of mass '*g*' contributed by each source to each individual sample as expressed in Eq 8. The residual e_{ij}, in Eq 8 is minimized using a least square approach (Paatero, 1997; Paatero and Tapper, 1994).

$$X_{ij} = \sum_{p=1}^{l} g_{ip} f_{pj} + e_{ij} \qquad \textbf{(8)}$$

The robustness of the model lies in the ability to weigh each individual data point using measured uncertainties and respective method detection limit (MDL) (Hopke, 2016). Missing values were replaced with associated geometric mean values. Results are constrained to have non-negative source contributions and profiles. The PMF solution minimizes the object function 'Q', based upon the uncertainties (*u*) associated with each sample using Eq 9.

$$Q = \sum_{i=1}^{m} \sum_{j=1}^{k} \left[\frac{X_{ij} - \sum_{p=1}^{l} g_{ip} f_{pj}}{u_{ij}} \right]^2 \qquad \textbf{(9)}$$

where, '*m*' and '*k*' are total number of samples and species, respectively.

This Q is the goodness of fits parameter and is used to check how well the model fit the given input data. Q_{robust}, means scaled residual is greater than 4 which is obtained after the exclusion of outliers, where as Q_{true} is calculated including all the data points. Along with the model Q_{robust} values, $Q_{theoretical}$ values are used to evaluate the model performance.

Categorization of quality of data was based on the signal to noise ratio (S/N) and the percentage of samples below MDL. The species S/N ratio >1 were categorized as 'strong', <0.2 as 'bad' and rest as 'weak'. Thereafter, uncertainties 'UC_{ij}' were calculated using Eq 10, unless C_{ij} is less than MDL_{ij}, where UC_{ij} is taken as 5/6th of MDL_{ij}.

$$\mathrm{UC}_{ij} = \sqrt{(0.1\times C_{ij})^2 + (0.5\times MDL_{ij})^2} \qquad \textbf{(10)}$$

Selection of number of factors/sources is completely user defined. Based on the user specified number of sources and iterations, the model runs and retrieves the results. Using the key tracers and knowledge of possible sources in a location, the adequate number of factors must be selected by the user and can discard if there any resolved factors are duplicate or have no physical sense or factors with unrealistic contributions.

Bootstrap analysis is used to assess the uncertainty of source profiles in order to minimize the randomized errors. F_{peak} is the parameter that can be used to rotate the obtained *g* and *f* matrices using the same Q value which is shown in Eq 11. But in general, in PMF it is not the rotation but known as the liner transformation of *g* and *f* matrices. Since, PMF results constrained to have non-negative source contributions and profiles, the rotation is possible if only the species in the new matrices are greater than zero. The solution is said to be unique, if there is no rotation possible,

$$g^* = gT, \qquad f^* = T^{-1}f, \qquad \textbf{(11)}$$

where, T is a non-singular matrix, *p* x *p*.

In EPA PMF v5.0, the base run for F_{peak} is automatically selected based on the lowest Q_{robust} value. After checking the appropriate number of boxes (-1.5 to 1.5) which signifies the desired strength of each F_{peak} run, five times the F_{peak} runs can be performed by the user. Positive F_{peak} value smears the matrix *g*, while sharpening *f* matrix and the negative F_{peak} value sharpens the matrix *f* and smear the *g* matrix.

3.5.3 CMB analysis

The US EPA's CMB v8.2 model uses source profiles and chemical species matrix as inputs (USEPA, 2004). It is necessary to have the prior information of pollution sources. CMB solves the mass balance equation expressed in Eq 8 and operates on the basis of effective variance-weighted least squares fitting (Hopke, 2016). The model requires uncertainty of each of the ambient data and source profile concentrations and the error is propagated in the iterative solution which determines the contributions and their uncertainties. Model provides many goodness of fit tests such as *t*-statistics, Chi square and R^2 to check the accuracy of the solution.

3.5.4 Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) analysis

Sources of moisture are important and their locations play a significant role to understand their influence on the study locations. Therefore, air parcel back trajectory analysis was used, which help us to find the source regions that contributes to the atmospheric pollutant concentrations (Huang et al., 2008). Data log of Global Data Assimilation System (GDAS) model using US National Oceanic and Air Administration (NOAA) Air Resource Laboratory's (ARL) HYSPLIT model (Draxler, 2011) used to plot the 5-day back trajectories with a starting height of 2000 m above ground level (AGL), which is a typical cloud height during rain events (Rao et al., 2016).

3.6 SUMMARY

To understand the dominant sources responsible for high concentration of PM in this region of India, wet deposition in an urban and dry deposition in three urban and two rural areas of Assam were collected. The chapter has been divided into five main sections: study area and sampling description, chemical characterization followed by the isotope analysis and regional deposition of size resolved PM in human respiratory tract and finally the source identification. Chemical characterization for both wet and dry depositions of PM were explained in detail along with the quality control techniques. The advanced laboratory testing techniques have been adopted for both chemical characterization and Isotope analysis for the rainwater samples collected. Details of each apparatus have been described in detail with the testing methodology. Source identification carried out for both wet and dry depositions of PM using PMF has been explained briefly.

CHAPTER IV – RESULTS AND DISCUSSION

4.1 RESULTS OF WET DEPOSITION OF PM IN AN URBAN LOCATION

4.1.1 Acidity and chemical composition

Total rainwater samples collected were 40, out of which 17 events were during monsoon, and remaining 23 samples belonged to the non-monsoonal period. The pH of these rain events varied from 4.59-5.99 with a mean of 5.20 (Fig 4.1). Further analysis indicated that 64% and 87% of the precipitation events during monsoon and non-monsoon season were acidic in nature, i.e. had pH less than 5.6. Reference pH of 5.6 is used as an indicator all around the globe to identify the acid rain events as, at this pH, cloud water will be in equilibrium with atmospheric CO_2 (Charlson and Rodhe, 1982).

Even though, most of the Indian cities reported alkaline rain events during the last decade, similar results 52% of acid rain events were reported by previous studies in Assam (Bhaskar and Rao, 2017; Kulshrestha et al., 2014). Such acid rain events in this region suggest the abundance of SO_2 and NO_x emissions and lack of buffering potential of the soil in this region (Kulshrestha et al., 2014). Also, heavy rainfall (mean annual precipitation of 1720 mm) and dense vegetation cover in this part of India make the soil containing components vulnerable from lifting into the atmosphere thereby leading to acid rain events (Bhaskar and Rao, 2017).

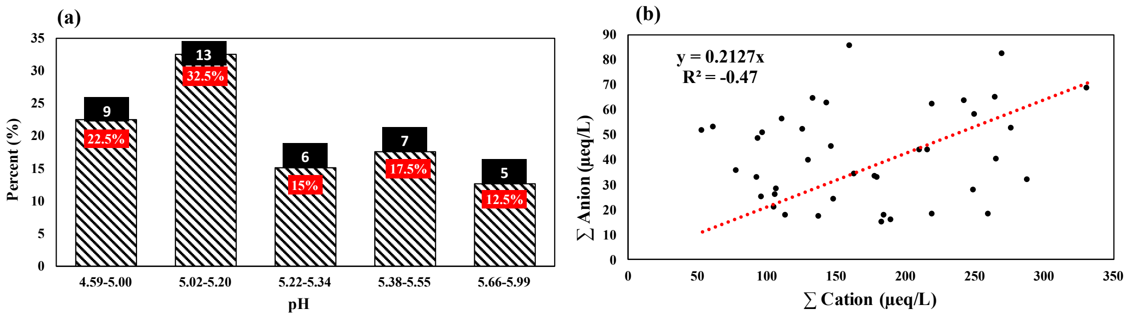


Fig 4.1 (a) Frequency distribution of pH and (b) ionic balance in the 40 rainwater samples collected from June 2016 to June 2017

The data of trace elements measured in the current study and studies from other UNESCO world heritage sites were shown in Table 4.1. The trend during the whole study period was: Zn (20%)>Sr (18%)>Mn (16%)>Fe (11%)>Cu (9%)>Pb (8%)>Ni> (6%)>Co (5%)>Cr (3%) and the concentration of Zn was maximum during non-monsoon while Sr being high in monsoon. Also, concentrations of the few trace elements such as Cu, Fe and Sr have increased by 65%, 42% and 67% respectively, during monsoon than non-monsoon. TOC concentrations in monsoon and non-monsoon seasons were similar.

The average ionic concentrations in the rainwater were in the order of SO_4^{2-} (65±32 µeq/L)> NO_3^- (60±31 µeq/L)> Cl^- (42±21 µeq/L)> Ca^{2+} (27±21 µeq/L)> K^+ (8±26 µeq/L)> F^- (7±10 µeq/L)> Na^+ (7±5 µeq/L)> Mg^{2+} (6±4 µeq/L). Table 4.1 also shows the comparison of ionic composition of the rainwater samples collected in the present study along with different UNESCO sites of the world. The acidic species (SO_4^{2-} and NO_3^-) were approximately similar to the other UNESCO sites. While in the case of crustal components (Ca^{2+}, K^+ and Mg^{2+}), the results were alike of the previous studies reported in this region but much less when compared to other sites.

Table 4.1 Comparison of chemical characteristics of rainwater measured from June 2016 to June 2017 at Guwahati, along with other UNESCO world heritage sites *(ions in µeq/L, trace elements in µg/L and TOC in mg/L*

Element	Guwahati, India[a]		Jorhat, India[b]	Petra, Jordan[c]	Philadelphia, USA[d]	Higashi–Hiroshima, Japan[e]	Sichuan, China[f]	Primorskaya, Russia[*]	Serpong, Indonesia[*]
	Monsoon	Non-monsoon							
F^-	0.32-52.79	0.47-12.16	1.80	NA	NA	NA	NA	NA	NA
Cl^-	14.39-78.36	5.78-80.93	7.70	80.60	NA	10.4	11.5-59.2	29.4	22.4
SO_4^{2-}	30.60-120.81	14.63-126.31	52.80	53.80	NA	14.7	6.8-1003.2	31.9	24.4
NO_3^-	9.31-120.24	30.07-118.61	39.20	35.70	NA	14.8	39.4-170.5	21.9	29.8
Na^+	0.65-15.00	1.96-28.04	10.10	75.60	NA	NA	6.2-34.8	23.9	8.32
K^+	0.39-7.31	0.77-20.34	6.30	18.40	NA	28	7.7-304.3	5.84	3.2
Ca^{+2}	8.98-49.40	8.98-81.09	41.80	71.40	NA	95	0.9-767.6	16.9	9.25
Mg^{+2}	1.25-16.25	1.25-15.00	9.80	62.30	NA	26	28.9-812.2	4.27	3.09
Co	40-180	40-200	NA	NA	0.51	NA	30.6-71.7	NA	NA
Cr	4-117	27-193	NA	NA	NA	NA	NA	NA	NA
Cu	40-600	20-240	NA	NA	0.56	0.62	NA	NA	NA
Fe	70-863	26-429	NA	NA	15.3	NA	NA	NA	NA
Mn	21-628	46-834	NA	NA	1.24	1.64	NA	NA	NA
Ni	13-234	24-730	NA	NA	1.12	0.26	NA	NA	NA
Pb	5-339	41-677	NA	NA	1.9	1.24	NA	NA	NA
Sr	55-876	69-464	NA	NA	NA	0.13	NA	NA	NA
Zn	23-674	153-809	NA	NA	5.16	4.77	NA	NA	NA
TOC	2.2-9.30	2.7-8.85	NA	NA	NA	NA	NA	NA	NA

Note:
NA: not available.
[a] Present study: 2016-17 sampling, India.
[b] (Kulshrestha et al., 2014): 2005-06 sampling, India.
[c] (Al-Khashman, 2005): 2002-04 sampling, Jordan.
[d] (Verry ES and Vermette, 1992): 1990-91 sampling, USA.
[e] (Takeda et al., 2000): Higashi–Hiroshima, 1995-97 sampling, Japan.
[f] (Qiao et al., 2015b): April 2010 to May 2011 sampling, China.
[*] (EANET, 2012): 2009-2010 sampling.

Therefore, higher contribution of acidic species indicated the influence of industrial sources, coal consumption and thermal power plants in and around the study area. Earlier studies reported the abundance of SO_4^{2-} in this region while Ca^{2+} being dominant in other parts of the country. About 78% of average ionic composition is formed by anions whereas remaining 22% is formed by cations. Maximum concentrations of the anions were observed during monsoon except for NO_3^- which was high in non-monsoon. Except Mg^{2+} (which was maximum in monsoon), all the other cations had high concentrations during non-monsoon. During monsoon, SO_4^{2-} was having the highest concentration among all. NF values for Ca^{2+}, Mg^{2+} and K^+ were 0.09, 0.01 and 0.03 respectively, during monsoon and 0.1, 0.01 and 0.07 respectively, during non-monsoon periods. Table 4.2 compares the NF of these ions of this study with the studies of UNESCO sites. When compared with those studies, except for Ca^{2+}, the other ions are having similar values as of those reported in other UNESCO sites, altogether showing poor neutralization. Such negligible neutralization capacity of alkaline species could be the reason for 76% of the acid rain events in this region (Das et al., 2005; Kulshrestha et al., 2014).

Table 4.2 Neutralization factor at different UNESCO world heritage sites

Area	**K^+**	**Ca^{2+}**	**Mg^{2+}**	**Reference**
Guwahati	0.046	0.092	0.007	This work
Jorhat	0.068	0.45	0.106	(Kulshrestha et al., 2014)
Jordan	0.206	0.79	0.69	(Al-Khashman, 2005)
Japan	0.96	3.20	0.89	(Takeda et al., 2000)
Russia	0.108	0.314	0.079	(EANET, 2012)
Indonesia	0.059	0.17	0.057	
China	0.082	1.90	0.50	(Qiao et al., 2015b)

4.1.2 Isotope analysis

Using all the event- based samples collected during the present study, the local meteoric water line (LMWL) in Guwahati (Fig 4.2 (a)) was established as:

$$\delta D\ (‰) = 8.40\ \delta^{18}O + 12.01 \quad (R^2 = 0.98) \qquad \textbf{(12)}$$

The slope and intercept of the LMWL are more or less similar to that of GMWL (Eq 5 in Section 3.3). Similar slopes indicate the raindrop formation in the tropical monsoon rainforest region of northeast India, under equilibrium conditions (Dansgaard, 1964) and unaffected by the evaporation effect (Craig, 1961). The average values of $\delta^{18}O$ and δD were -4.11‰ and -23.41‰ during monsoon, which were much less than the average values during non-monsoon i.e., -0.80‰ and 5.60‰. This suggests that the occurrence of isotopically enriched rain events during non-monsoon, which was also observed in a study conducted in southern India (Srivastava et al., 2014). The d-excess values, which serves as an essential tool to identify the moisture source, and if d-excess>10‰, it implies that the moisture source is typically from the continental region, i.e. evapotranspiration and nominally from the ocean (Wirmvem et al., 2017). In the present study, the d-excess values for monsoon and non-monsoon were 9.47 and 12.14‰ respectively.

Further, in order to understand the seasonal influence on the isotopic composition of precipitation, the $\delta^{18}O$ vs d-excess was plotted as shown in the Fig 4.2 (b). The distribution shows two distinct different groups of data points having two very different trend lines, indicating sample events belonged to two different groups, monsoon and non-monsoon. A higher slope in the $\delta^{18}O$ vs d-excess space represents a strong raindrop evaporation undergoing during the non-monsoon season and a reduced slope representing low evaporation as a result of increased humidity during the monsoon season.

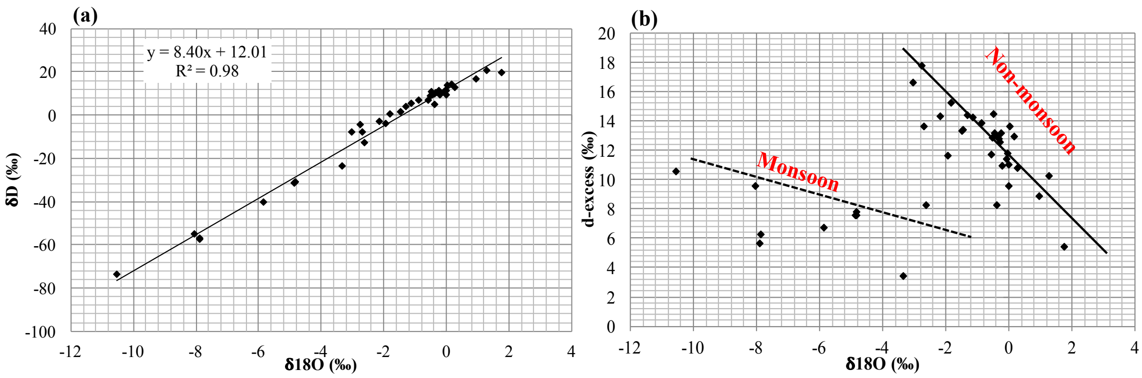
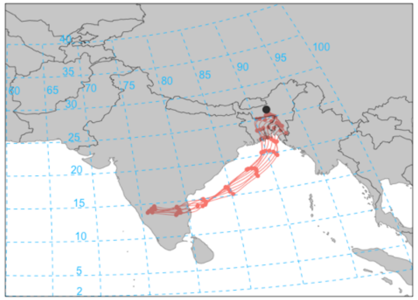


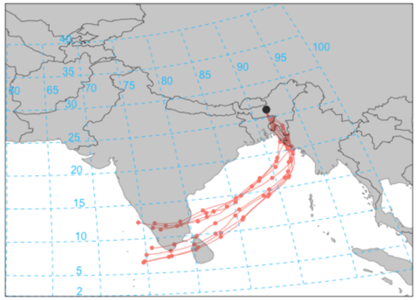
Fig 4.2 (a) Local meteoric water line for the sample events collected during June 2016 – June 2017 (b) seasonal influence on isotopic composition using d-excess (‰) and **δ**^{18}O (‰) .

Fig 4.3 shows the HYSPLIT trajectories during monsoon season, having d-excess<10‰ for most of the individual rain events. For few of the rain events during monsoon, have d-excess>10‰ which could be due to feeding of secondary water vapor into the cloud system (Chakraborty et al., 2016). While Fig 4.4 shows the trajectories during non-monsoon having d-excess>10‰. Therefore, the trajectories originated from majority of the individual rain events indicated that the maximum contribution from ocean during d-excess<10‰ i.e. monsoon and water-inland during d-excess>10‰ i.e. non-monsoon.

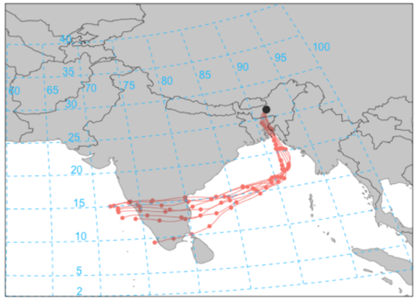
Therefore, the isotope analysis which is used to identify the geographic origin of the moisture, revealed that the significant amount of rainwater during non-monsoon originated from the water inland i.e. long range transport while during monsoon the moisture is transported from the ocean which is inline with HYSPLIT analysis.



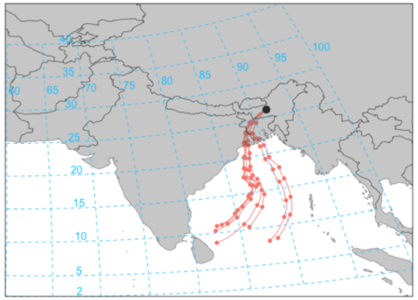
16-6-2016(d-excess=7.53‰)



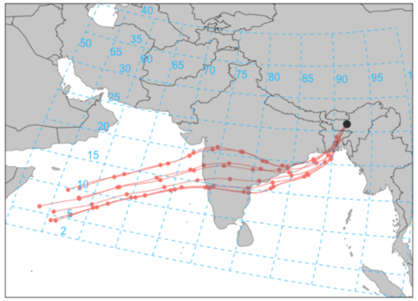
17-6-2016(d-excess=3.40‰)



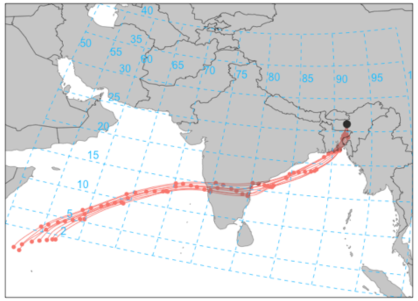
18-6-2016(d-excess=7.77‰)



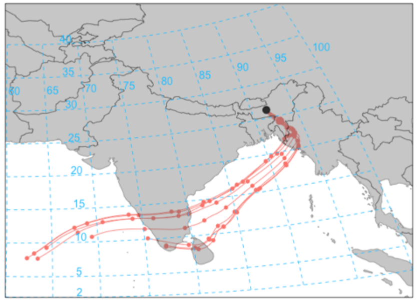
23-6-2016(d-excess=7.58‰)



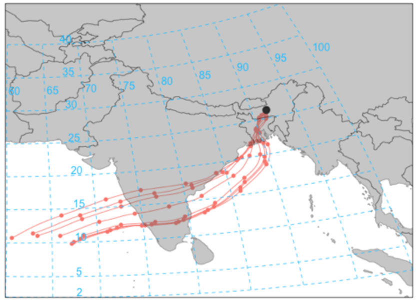
4-7-2016(d-excess=6.75‰)



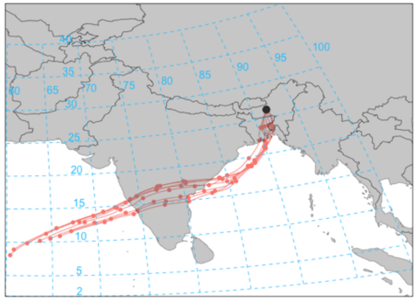
6-7-2016(d-excess=5.65‰)



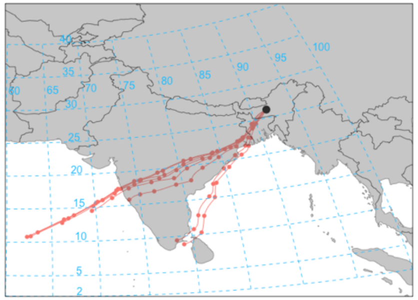
8-7-2016(d-excess=6.22‰)



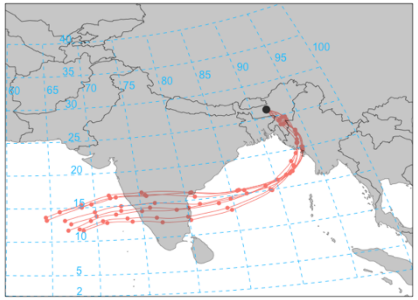
14-7-2016(d-excess=9.52‰)



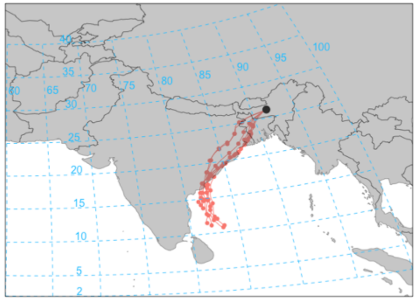
16-7-2016(d-excess=10.55‰)



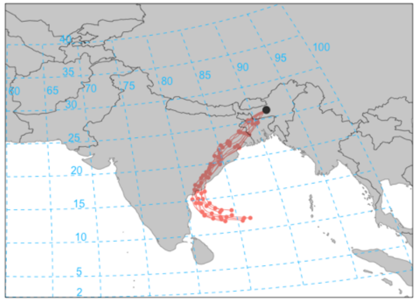
19-7-2016(d-excess=11.02‰)



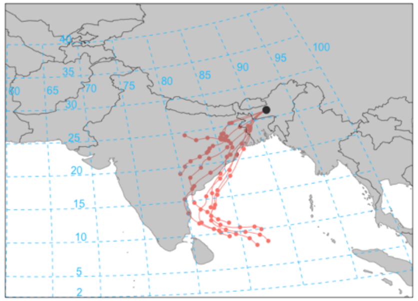
4-8-2016(d-excess=11.67‰)



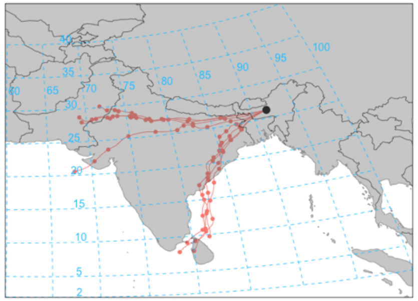
5-5-2017(d-excess=13.36‰)



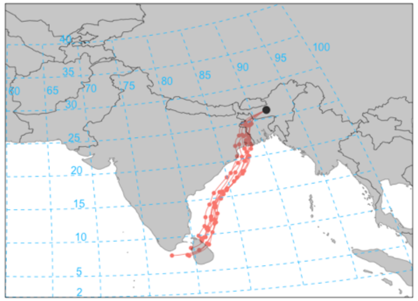
31-3-2016(d-excess=8.27‰)



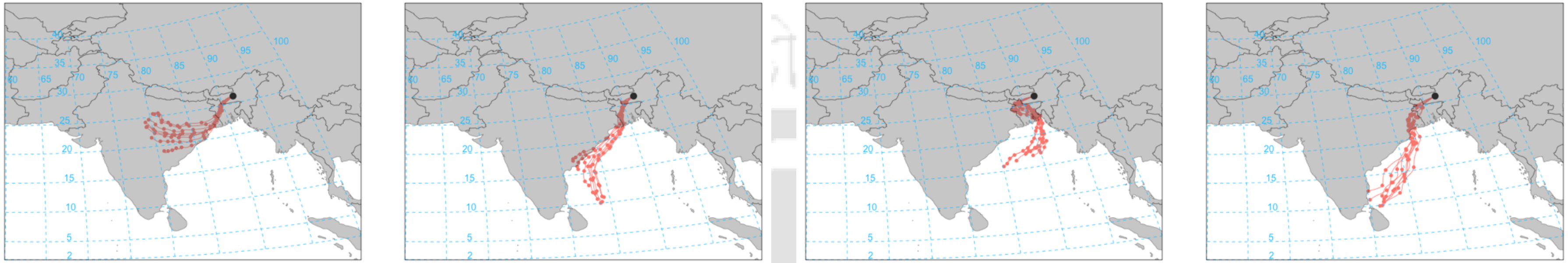
3-4-2017(d-excess=8.26‰)



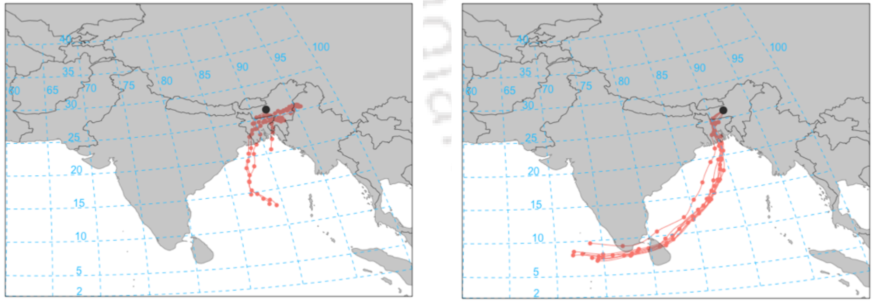
25-4-2017(d-excess=9.57‰)



29-4-2017(d-excess=5.45‰)



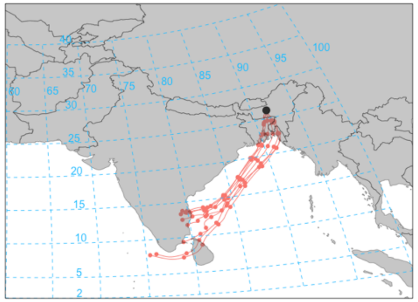
9-5-2017(d-excess=13.18‰) 10-5-2017(d-excess=10.23‰) 18-5-2017(d-excess=11.80‰) 22-5-2017(d-excess=8.83‰)



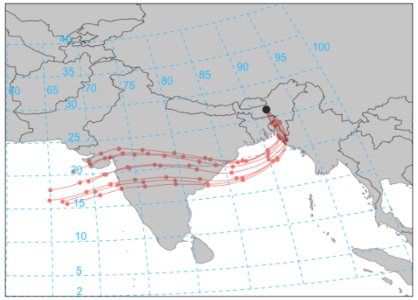
28-5-2017(d-excess=11.22‰) 3-6-2017(d-excess=11.42‰)

Fig 4.3 The 5-day back trajectories developed using HYSPLIT for the individual rain events in monsoon (22 events)

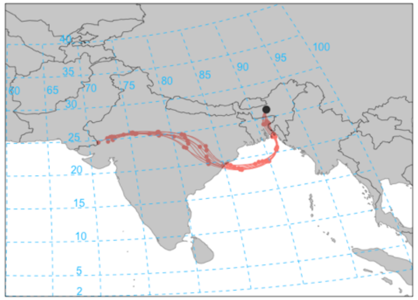
26-8-2016(d-excess=13.88‰)



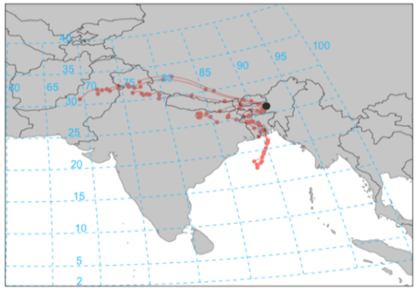
3-9-2016(d-excess=12.94‰)



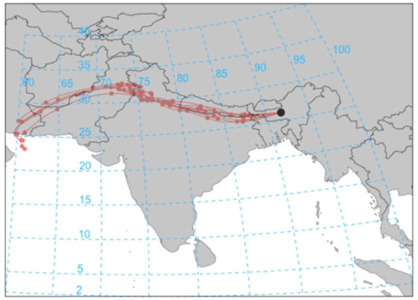
5-9-2016(d-excess=13.15‰)



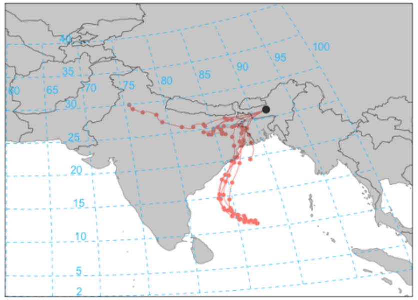
8-9-2016(d-excess=14.39‰)



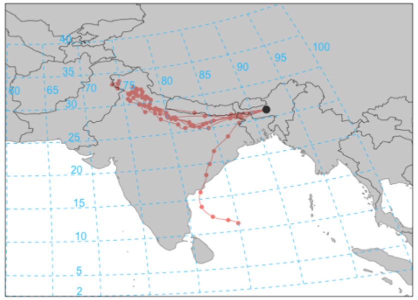
27-3-2017(d-excess=17.77‰)



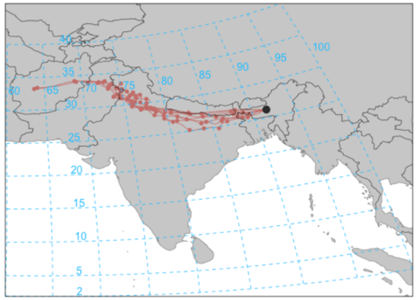
1-4-2017(d-excess=14.33‰)



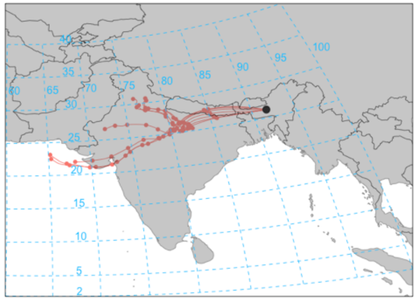
2-4-2017(d-excess=16.58‰)



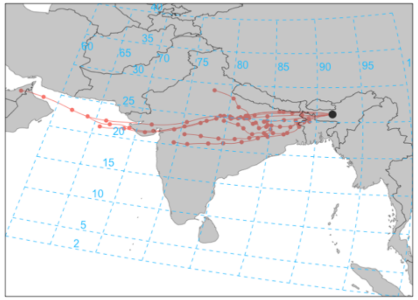
4-4-2017(d-excess=13.61‰)



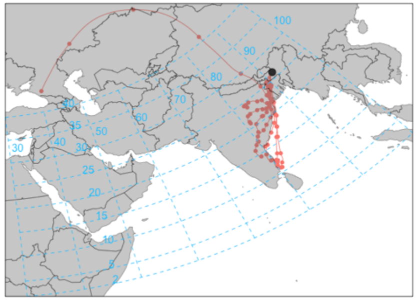
5-4-2017(d-excess=15.20‰)



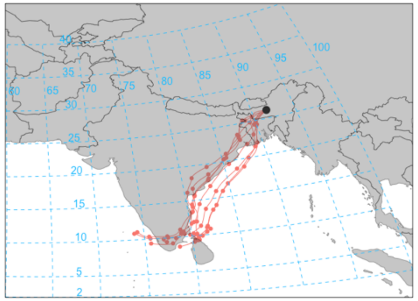
7-4-2017(d-excess=14.26‰)



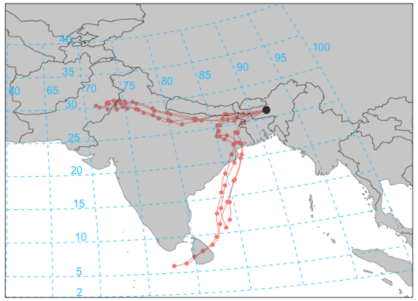
8-4-2017(d-excess=12.89‰)



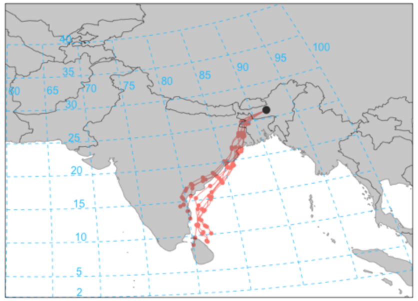
19-4-2017(d-excess=12.82‰)



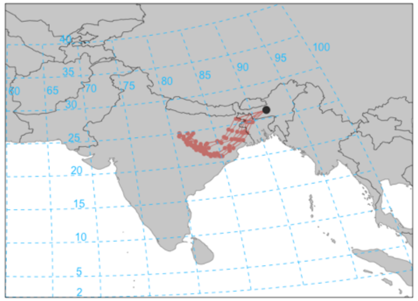
22-4-2017(d-excess=10.75‰)



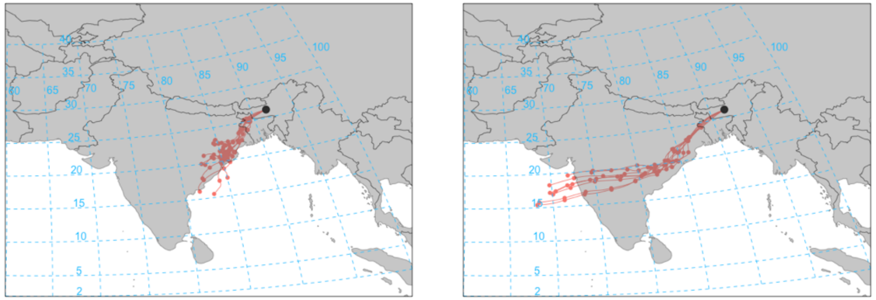
24-4-2017(d-excess=11.66‰)



1-5-2017(d-excess=13.32‰)



20-5-2017(d-excess=12.55‰)



21-5-2017(d-excess=13.63‰)   2-6-2017 (d-excess=12.81‰)

Fig 4.4 The 5-day back trajectories developed using HYSPLIT for the individual rain events in non-monsoon (18 events)

4.1.3 Source identification

The source apportionment results carried out using EF analysis for different elements are displayed in Fig 4.5. It is evident that the elements except Fe, other elements like Cr, Ni and Sr have the mean EF's >3. EF of Fe is close to unity meaning it is of geological origin. Whereas, all the other elements i.e. Pb, Zn, Co and Cu have the EF's >10, indicating the influence of anthropogenic sources. Therefore, EF analysis in the present study served as an effective tool in identifying the impact of elemental concentrations by human activities other than natural sources.

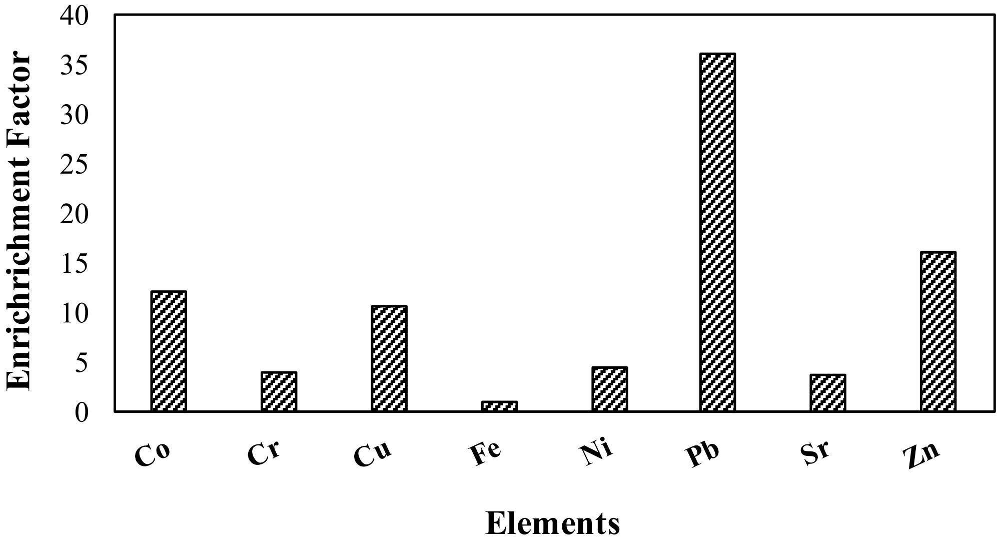


Fig 4.5 Distribution of enrichment factors for the trace metals with Mn as reference element

EPA PMF v5.0 using the dataset of 40 samples were employed and 4 to 7 factor solutions were tested wherein, mixed and unidentifiable sources were obtained. Hence 5-factor solution with Fpeak of -0.5 yielded the most reasonable source apportionment results. Also, in the present study the elemental forms of K, Mg, Na, Cl and Ca are not included since their ionic forms were taken into consideration in order to avoid multiple counting of mass.

Fig 4.6 shows the source profiles and its associated percentage of species in a source, which is the median value from bootstrap runs.

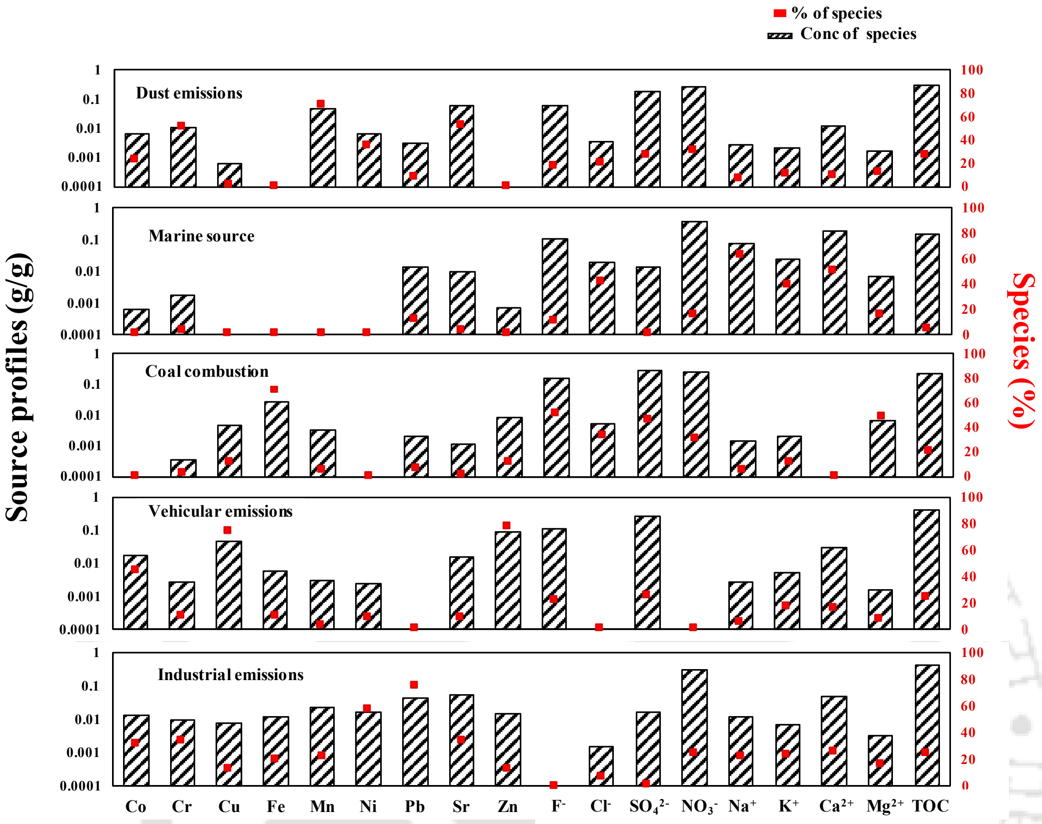


Fig 4.6 Source profiles resolved by EPA PMF v5.0 model for the rainwater samples collected during the study period

Dust emissions: This factor explained 26% of the total variance with high relative abundances of Mn (69%), Sr (52%) and Cr (51%). These species are tracers of resuspended dust and crustal sources and are largely reported by many researchers (Crilley et al., 2017; Xue et al., 2010; Zhao et al., 2011). Since, the study area is close to the highways, it is influenced by the abrasive emissions emitted from the highway surfaces.

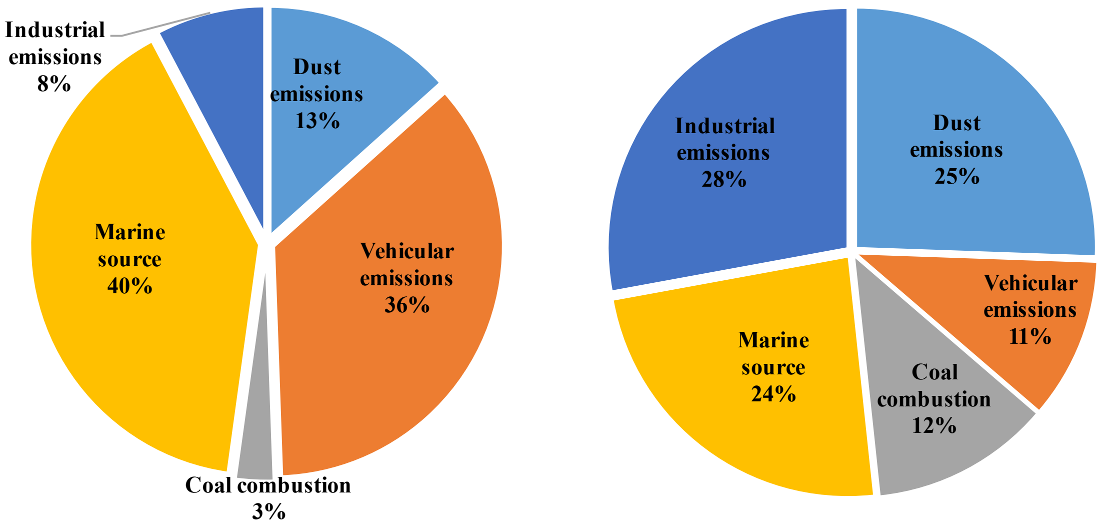
Marine source: This source has maximum (28%) contributions to measured constituents in rain water. High loadings of Na^+ (61%), Cl^- (40%), Ca^{2+} (49%) and K^+ (38%) characterized this source (Kothai et al., 2011; Police et al., 2016; Sharma et al., 2014). The general use of K^+ as an indicator to biomass burning and Cl^- as coal combustion tracer in Indian studies, leads to ambiguity however, the combination of Cl, Na, K, Ca should provide a better signature.

Coal combustion: This factor is mainly characterized by significant levels of Fe (69%), F^- (51%), SO_4^{2-} (45%), NO_3^- (33%) and Cl^- (32%) and accounts for 9% to the rainwater constituents. Based on the previous studies, these species were reported as tracers for coal combustion and indicating that the surrounding regions have been influenced by the coal consumption activities (Machado et al., 2015; Qiao et al., 2015b; Sharma et al., 2016b; Tiwari et al., 2016).

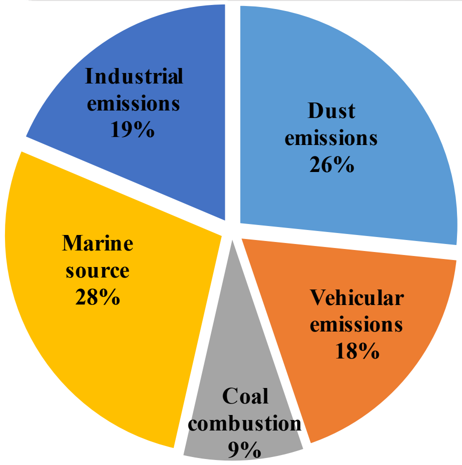
Vehicular emissions: This factor accounted for 18% and contributed to high proportions of Zn (77%), Cu (74%), Co (44%) and TOC (25%). Earlier studies reported that Zn, Cu, Co and TOC are the elemental tracers and reliable markers for the vehicle exhaust emissions (Chakraborty and Gupta, 2010; Dadashazar et al., 2019; Sharma et al., 2014). As this area experiences a huge traffic flow round the year, hence this source is expected.

Industrial emissions: This factor indicated high loadings of Pb (75%), Ni (57%), Sr (34%), Cr (34%) and Co (32%) and contributed 19% of the rainwater constituents. Previous studies suggested that these elements are mainly emitted from industries like steel, electroplating, metallurgy, foundry (metal casting), machinery and battery, etc. and are used as key markers to identify industrial emissions (Edwards, 1997; Krishnamurthy et al., 2014; You et al., 2017).

Fig 4.7 shows the variation of source contributions in monsoon, non-monsoon and all samples (combined). Marine source (28%) and dust emissions (26%) were the main sources during whole study period (panel (c)). In monsoon samples (when d-excess<10‰), marine source (40%) and vehicular emissions (36%) were the major contributors (panel (a)) indicating the probable moisture source region is from ocean. While on the other hand, the contributions from the marine source decreased by 16% and industrial emissions increased by 20% in the rain events occurred during non-monsoon periods, as the rain water droplet was more of in-land origin.



(a) Monsoon **(b) Non-monsoon**



(c) Combined

Fig 4.7 Source contributions obtained using EPA PMF v5.0 model during (a) monsoon (b) non-monsoon (c) and combined

4.1.4 Summary

The wet deposition of PM analyzed revealed a year-long acid rain events ($pH<5.6$) in this region. Some of the major ions, trace metals and TOC measured in the rainwater samples indicated the low concentrations of crustal species with neglible buffering capacity of acidic species. The isotope analysis in conjunction with back trajectory analysis conducted, could identify that the significant source of rainwater during non-monsoon was originated from water inland, while during monsoon the moisture transported from the ocean.

Also, enriched isotopic composition in non-monsoon season (average Deuterium excess (d-excess) of 9.47 ‰), and isotopically depleted rains were during monsoon period (average d-excess of 12.14 ‰) was observed during the study period. These results bolster the conclusions from the US EPA's PMF analysis that showed higher contributions from marine sources in monsoon and industrial sources in non-monsoon seasons, respectively. Also, the Enrichment Factor (EF) analysis identified the enriched contributions of Pb and Zn which are typically of anthropogenic origin suggesting that this region needs the implementation of scientifically based control strategies so as to regulate the anthropogenic activities which lead to severe acid rains in this locality.

4.2 RESULTS OF DRY DEPOSITION OF PM IN FIVE LOCATIONS OF ASSAM

4.2.1 Seasonal variation of PM_{10} and $PM_{2.5}$ concentrations

Fig 4.8 illustrates the seasonal variations of the average concentrations of PM_{10} and $PM_{2.5}$ in all the selected locations along with the standard errors. Two groups of sites, i.e. urban (S1, S2 and S3) and rural (S3 and S4) each with varied concentrations during three seasons, can be distinguished. During this study, the 24 hour National Ambient Air Quality Standard (NAAQS) for PM_{10}, i.e. 100 µg/m^3, was exceeded in 78% of the samples and for $PM_{2.5}$, i.e. 60 µg/m^3, was exceeded in 85% of the samples.

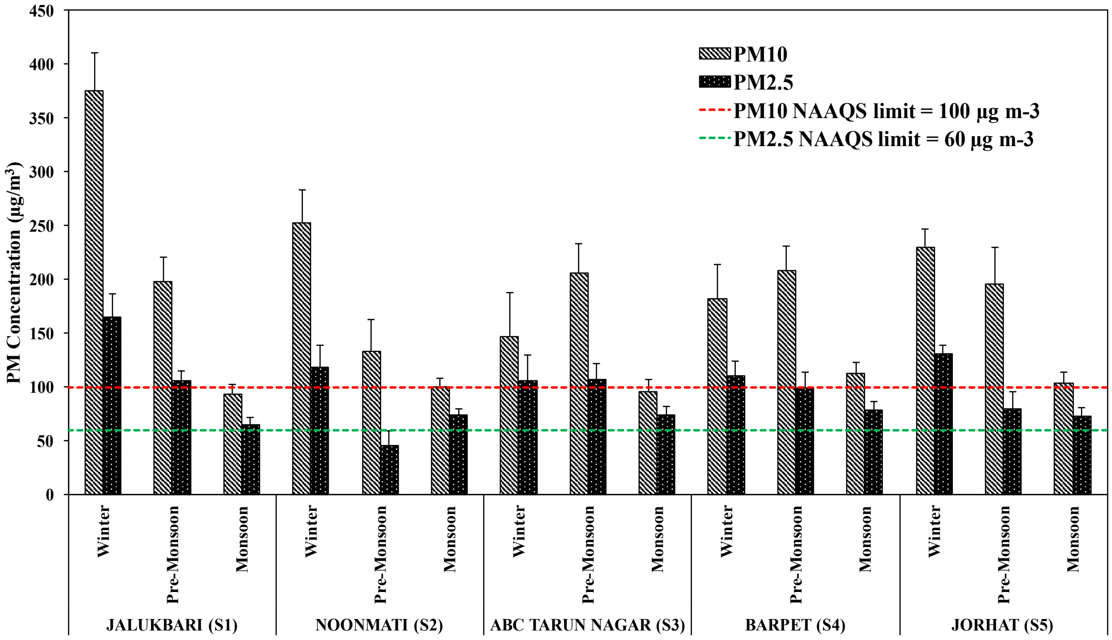


Fig 4.8 Seasonal variation of PM_{10} and $PM_{2.5}$ concentrations in all the five locations

Table 4.3 suggests the variability of concentrations of $PM_{2.1}$ in the present study and the studies elsewhere which used 8-stage cascade impactor. The concentrations are comparable with the studies within India and relatively higher than those reported in China.

Table 4.3 Comparison of concentration of fine PM in the present study along with the studies elsewhere which used 8-stage cascade impactor

Site	Year of sampling	$PM_{2.1}$ Mass	Type	Reference
S1	Jan’2018-Dec’2018	111±12.6	Residential/ Ring road	Present study
S2		79±13.56	Industrial	
S3		95±15.31	Residential	
S4		96±11.90	Rural/ background	
S5		94±10.91	Mining	
Chandrapur, India	May’2010-Feb’2011	54.87	Mining	(George et al. 2017)
Ghugus, India	May’2011-Feb’2012	79.50	Mining	
Okha, India	Sep’-Oct’2010	30.46	Rural/ Background	
Delhi (road dust), India	Sep’-Oct’2005	154.56	Urban	
Delhi (vehicle), India		160.22	Urban	
Site 1, Kolkata, India	Dec’2003-Feb’2004	96-355	Industrial/ Residential	(Nag et al. 2005)
Site 2, Kolkata, India		116-363	Commercial	
Site 3, Kolkata, India		99-263	Commercial	
Rajim*, India	Jul’2012-Jun’2013	101±55.7	Rural	(Nirmalkar et al. 2016)
Raipur*, India	Jul’2009-Jun’2010	150±78.6	Urban	(Deshmukh et al. 2013a)
Durg*, India	Jul’2009-Jun’2010	135±76.2	Industrial	(Deshmukh et al. 2013b)
Nagpur, India	2008-2009	136.7	Residential/ commercial	(Pipalatkar et al. 2012)
Gulou, China	May’2010-Apr’2011	55.1±36.3	Urban	(Chen et al. 2015)
Pukou, China		64.8±40.2	Sub-urban	
Gulou, China	Jul’2013-Sep’2013	50±12.8	Urban	(Chen et al. 2017)
	Jul’2014-Sep’2014	34.6±17		

*Note: The cut size of fine PM is 2.5 µm

Concentrations were found to be higher during winter and pre-monsoon while being the least in the monsoon period. The prevailing weather conditions in winter, such as calm winds and low mixing layer heights prevent the dispersion of PM resulting in the accumulation near the surface. This probably led to higher concentration in winter than monsoon. The PM in this region accounts for higher concentrations which was also reported recently by centre for northeast studies and policy research (C-NES) which claimed that this region ranks high in the pollution charts, in competence with metros like Delhi and Kolkata (https://www.c-nes.org/1461/c-nes-report-on-air-pollution-released/). As indicated earlier, PM_{10} and $PM_{2.5}$ concentrations exceeding NAAQS limit, the present study therefore strongly suggests the importance of fine PM due to their distribution of 50% in total mass of PM_{10}.

4.2.2 Fractional deposition of PM in the human respiratory tract

The average deposition fraction in NOPL, TB and P in respective size range is shown in Table 4.4.

Table 4.4 Average regional deposition fraction in respective size range

Size range	NOPL	TB	P
Back up filter (0-0.4)	0.04890	0.10576	0.37854
Stage 7 (0.4-0.7)	0.15133	0.01402	0.11740
Stage 6 (0.7-1.1)	0.27709	0.02094	0.14773
Stage 5 (1.1-2.1)	0.45399	0.03181	0.18953
Stage 4 (2.1-3.3)	0.64942	0.04281	0.19708
Stage 3 (3.3-4.7)	0.78460	0.04905	0.16101
Stage 2 (4.7-5.8)	0.86241	0.05168	0.11598
Stage 1 (5.8-9.0)	0.91570	0.05188	0.05654
Stage 0 (9.0-10)	0.92740	0.04985	0.01646

Fig 4.9 shows the fractional deposition of size resolved PM in the human respiratory tract for all the selected locations during winter, pre-monsoon and monsoon. Significant

difference between mass concentration and fractional deposition of PM in the human respiratory tract, in all the sites was noticeable. For example, in the case of urban sites, in winter, the concentration of PM_{10} was high in S1 when compared to other sites. But the fractional deposition of PM in S3 in P region was 0.61, which is 1.7 times higher than S1. Similarly, in the case of rural sites, in pre-monsoon, although the concentration of PM_{10} was high in S4 than S5, but, the fractional deposition in TB region was 0.06 for S5 which is 63% less than S4. Overall, the total fractional deposition of PM was high in NOPL, where the coarse particles were largely deposited than TB and P in most of the sites. This could be due to the deposition fractions of respective size ranges for NOPL, TB and P (Fig 3.5). Similar observations were reported by the recent study conducted in China, where deposition mass was estimated (Liu et al., 2019). The chemical composition and their deposition variation in various regions of respiratory tract were presented in Annexure D.

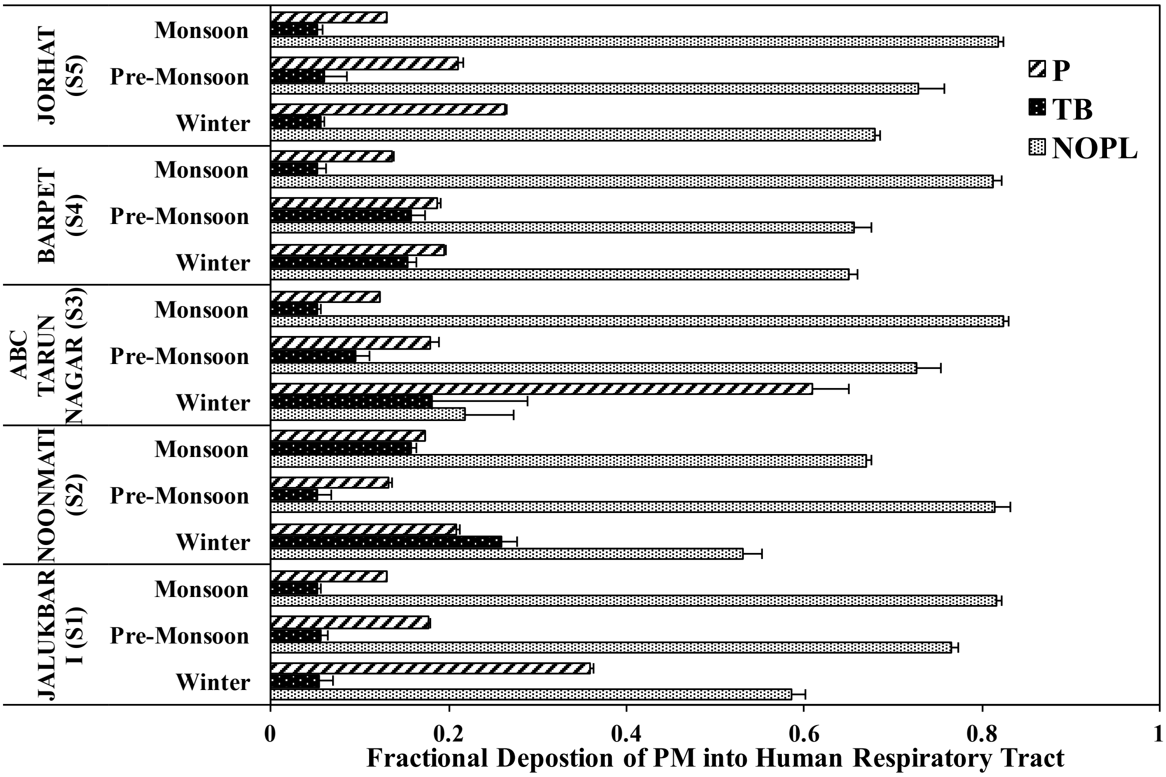


Fig 4.9 Fractional deposition of PM in the sampled locations during all the seasons

4.2.3 Source identification

For the size segregated aerosol measurements, a speciated matrix of 21 species (Ca^{2+}, NH_4^+, Na^+, K^+, Mg^{2+}, F^-, Cl^-, NO_3^-, and SO_4^{2-}, Al, Cd, Co, Cu, Cr, Fe, Mn, Ni, Pb, Sr, Zn and TOC) has been included as input in the EPA PMF v5.0 model along with the estimated uncertainties. The number of valid samples in each location per season were 126 which accounted for 378 number of samples in each of NOPL, TB and P regions. 20 base runs were selected to identify the probable number of factors/sources, maintaining good agreement between Q and $Q_{theoretical}$. Q_{true} was within 1.5 times of Q_{robust}. Various extra modelling uncertainties were used to obtain the reliable outcomes, which are statistically and physically interpretable. F_{peak} of optimum value with 100 bootstrap runs and minimum R^2 of 0.6 was chosen to test the uncertainty, in order to obtain the efficient solution to ensure better physical interpretation of the sources. Solutions of 6-8 factors were carefully examined based on the key tracer species. Detailed description of the PMF factor profiles for S1, S2, S3, S4 and S5 in NOPL, TB and P regions were given below. Fig 4.10 shows the source profile of S1 location and its associated percentage of species in a source, which is the median value from bootstrap runs for all the three regions of human respiratory tract.

Vehicular emissions: This factor accounted for 27%, 38% and 39% in NOPL, TB and P respectively. High proportions of Cu, Zn, Ni, Pb, Mn and TOC were contributed to this factor. Earlier studies reported that Zn, Cu, Ni, Mn and TOC are the elemental markers for the vehicular emissions (Chakraborty and Gupta, 2010; Dadashazar et al., 2019; Sharma et al., 2014). The proximity to major national highways i.e. NH 31 at north and NH 37 at south, this area acts as quadruple road junction which is also signal-free intersection, enabling free movement of traffic in all the four directions in and through the Guwahati city all-round the year (https://www.telegraphindia.com/states/north-east/nhai-to-deck-up-jalukbari/cid/1408612). Thus, this source is highly expected.

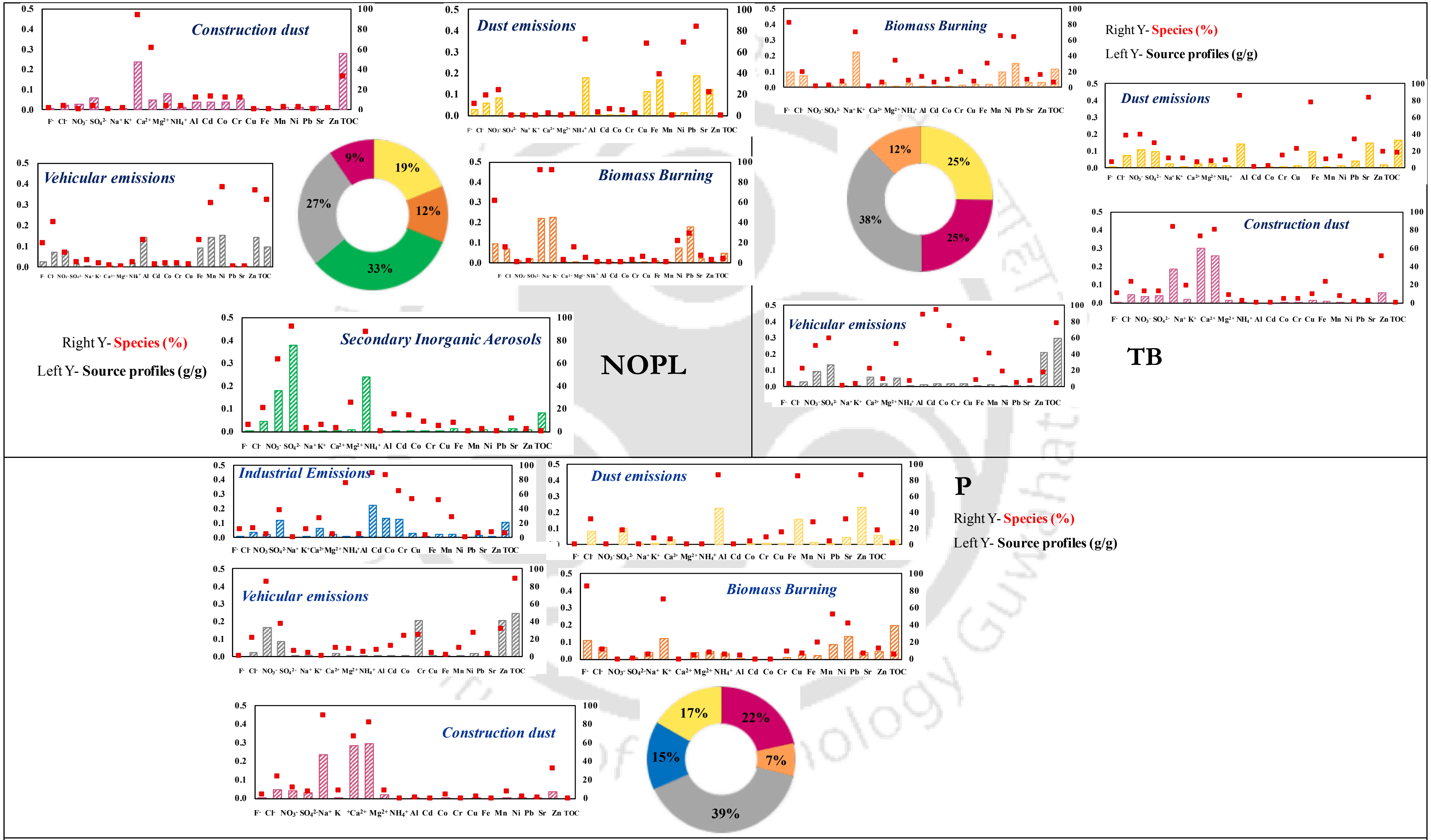


Fig 4.10 Source profiles resolved by EPA PMF v5.0 model for samples collected in S1 during the study period

Construction dust: This factor is mainly characterized by high loadings of Ca^{2+}, Na^{+} and Mg^{2+}, which are assumed to be originated from the construction activities. Ca^{2+}has been one of the major representative tracer species for the construction dust as it is the main element in cement or plaster (Kim et al., 2004). This factor accounted for 9%, 25% and 27% in NOPL, TB and P respectively. The proportion of particles less than PM_{10} in cement dust are very high, even though their size generally from 3-50 µm (Abdul-Wahab, 2006). This could be the probable reason for the maximum contribution in TB and P than NOPL. However, this area is mainly notable for the cement factories and on-going construction activities all throughout the year, of which many projects were undertaken by government such as National Highways Authority of India (NHAI). Therefore, this source is obvious and was reflected in all the three regions of respiratory system i.e. NOPL, TB and P, due to the large emissions of pollutants from the construction activities in this area.

Dust emissions: This factor is represented by the significant levels of Al, Fe, Mn, Zn contributing 19% in NOPL, 25% in TB and 17% in P regions. According to the previous studies, Al and Mn are the important markers for the dust emissions (Crilley et al., 2017; Xue et al., 2010; Zhao et al., 2011). The wind-driven airborne dust from surface soils would have resulted in the considerable emissions of this factor.

Biomass burning: This factor accounted for relatively minimal contributions of 12%, 12% and 7% in NOPL, TB and P respectively. Major proportions of K^{+} and F^{-} were contributed to this factor. There have been many studies in the past suggesting that K^{+} as clear indicator of biomass burning. (Sharma et al., 2014). It is a known fact that owing to socio-economic reasons, biomass is a widely used energy source in northeast India wherein the implications of it have resulted in the nominal contributions of biomass burning in this location.

Secondary inorganic aerosols: This factor is interpreted as secondary aerosol as it has high loadings of SO_4^{2-} (89%), NO_3^-(64%) and NH_4^+(87%) accounting for 33% in NOPL region having no effect in TB and P regions. Many source apportionment studies have reported SO_4^{2-}, NO_3^- and NH_4^+ as marker species in the past (Srimuruganandam and Shiva Nagendra, 2012). The formation of secondary aerosols is due to the chemical transformation. These secondary ions are derived from gas to particle conversion processes involving photo-chemical reaction of gaseous precursors such as SO_2 and NO_x which are largely emitted from local and regional sources (Pandolfi et al., 2011).

Industrial emissions: This factor has high levels of Cd (88%), Co (87%), Cr (60%) and Cu (55%) contributing 15% to total PM concentration in P region. Earlier studies reported that Cd, Cu, Co and Cr are the indicators of the industrial emissions as these elements are greatly used in various industries like machinery, battery and electroplating purposes (Taghvaee et al., 2018). Location of Amingaon (south) and Boragaon (north) industrial regions at an aerial distance of 3.5 km and 4 km, respectively in this area could be the possible reason of this source.

Fig 4.11 shows the source profiles resolved using PMF model at S2 location. The first source was named as vehicular emissions due to considerable levels of Cu, Fe and Zn and has contributed 20%, 18% and 24% to total PM in NOPL, TB and P regions, respectively. Due to the increased traffic in Guwahati, the capital city of Assam, the hierarchy based arterial road network system is operated with inclusion of major corridors to enable smooth flow of traffic in the city. Out of the major corridors, S1 and S2 form an important radial corridor which carries the substantial amount of traffic (https://gmda.assam.gov.in/portlet-innerpage/transport-2025). Therefore, the emissions due to heavy traffic in this area could

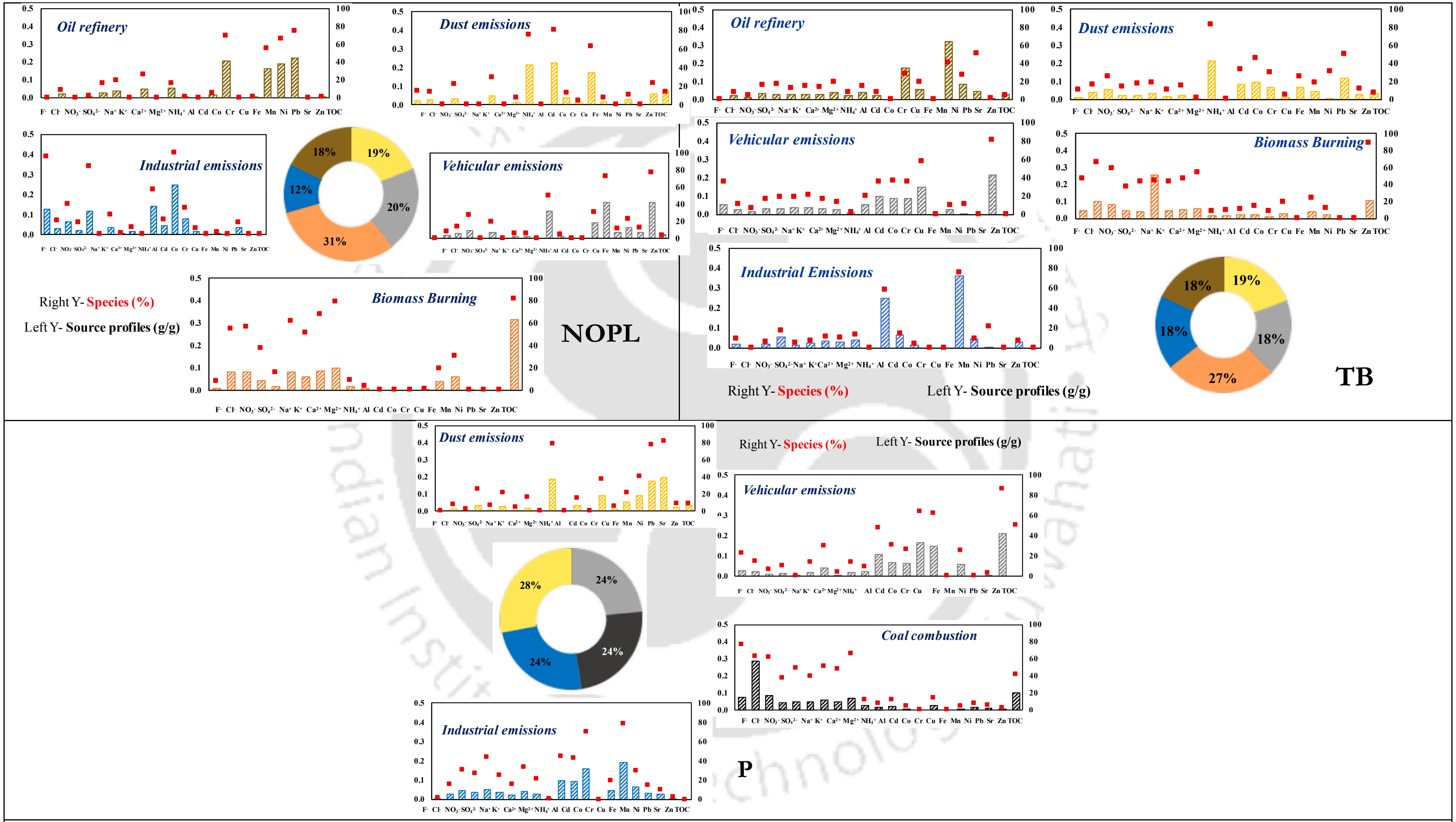


Fig 4.11 Source profiles resolved by EPA PMF v5.0 model for samples collected in S2 during the study period

reflect in the occurrence of this factor in all the three regions of human respiratory tract. The second source has accounted for 19%, 19% and 28% in NOPL, TB and P regions, respectively with high loadings of Al, Mn, Fe and Cd and is therefore treated as dust emissions. Significant levels of Cd, Co, Cr and Cu have been attributed to the third source named as industrial emissions, which has contributed 12% in NOPL, 18% in TB and 24% in P regions. Unvaried contribution of oil refinery with 18% each in NOPL and TB regions has been observed as fourth source. High proportions of Ni, Pb, Cr and Sr were noticed where Ni and Pb were generally termed as key tracers of oil refinery by the previous studies (Negi et al., 2002). As this area is well known for its existence of Guwahati refinery which is the first and foremost oil refinery undertaken by Indian oil company in 1962 (https://en.wikipedia.org/wiki/Noonmati). So, its influence on direct emissions was speculated from the results. Finally, the last source was named as coal combustion as the loadings of Cl^-, F^-, Na^+, SO_4^{2-} and NO_3^- were observed and this factor accounted for 24% in NOPL region alone. Literature studies directly linked the elevated levels of Cl^- as one of the main tracers for coal combustion (Zong et al., 2016). This region of northeast India is rich in fossil fuels like coal, oil and gas which are generally used for energy generation in the thermal power plants (https://assam.gov.in/en.). This factor is thus to be expected.

Fig 4.12 shows the resolved source profiles and contributions at an urban location S3. Overall, 5 to 6 sources in all the three regions i.e. NOPL, TB and P were noticed. In case of NOPL, contributions of construction dust (23%) were high its influence is confined in this region alone. Whereas, the stable contributions were shared among secondary aerosols (17%), biomass burning (17%) and dust emissions (18%). Also, the minimal but considerable emissions from industries (13%) and vehicular traffic (12%) were observed. While on the other hand, in TB region, biomass burning (27%) was the chief contributor

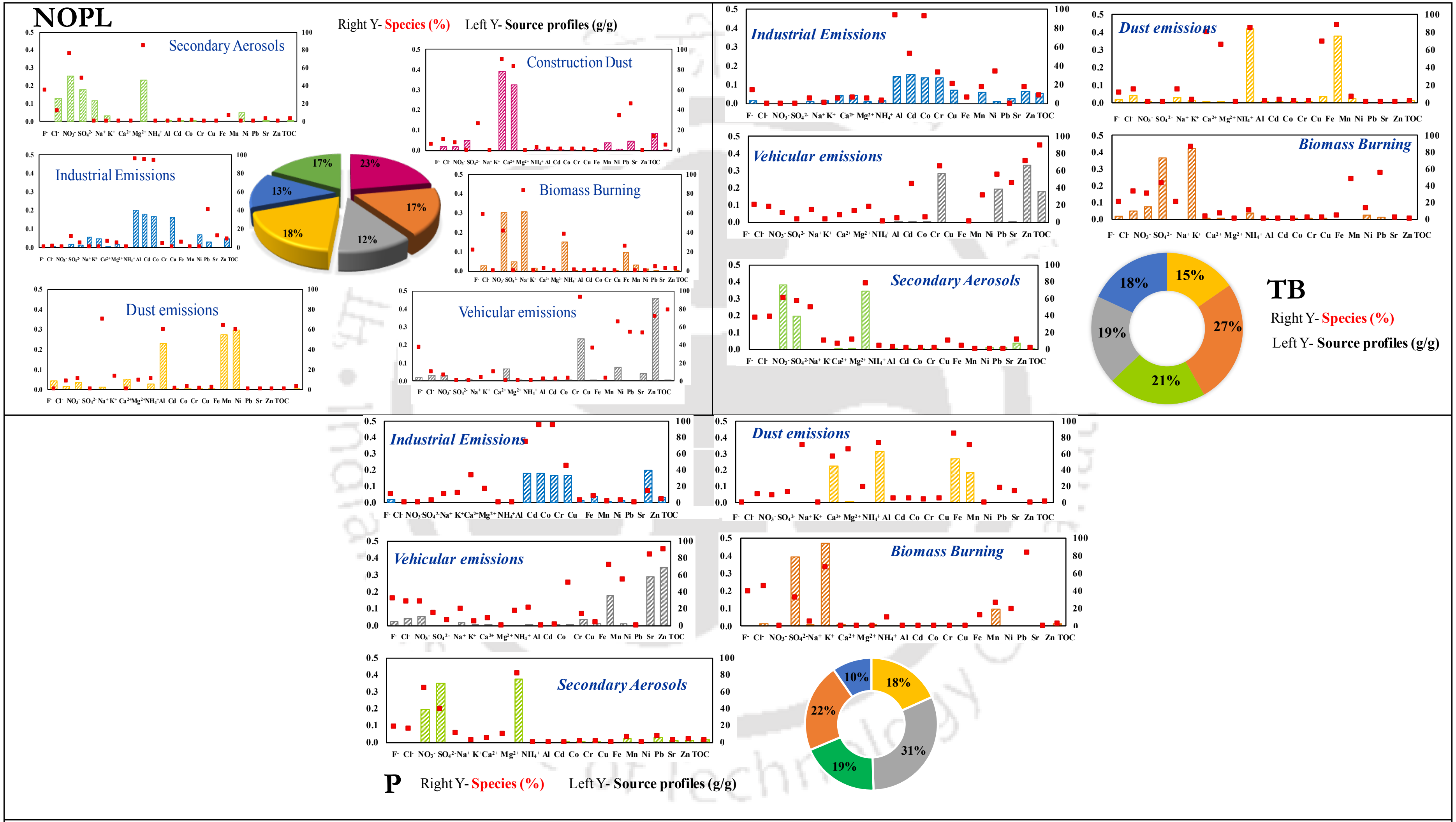


Fig 4.12 Source profiles resolved by EPA PMF v5.0 model for samples collected in S3 during the study period

along with the second highest source contributor as secondary aerosols (21%). Moderate contributions of dust emissions (15%), industrial emissions (18%) and vehicular emissions (19%) were inspected. Further, in P region, vehicular emissions being the dominant source elevated by 2.5 and 1.6 times than NOPL and TB regions contributing 31% to the total PM mass. However, the industrial emissions (10%), biomass burning (22%) and secondary aerosols (19%) were decreased by 1.8, 1.2, 1.1 times, respectively than TB. While, the emissions of biomass burning and secondary aerosols increased by 1.3 and 1.1 times than NOPL with marginal decrease of industrial emissions by 1.3 times. The last source i.e. dust emissions was accounted for 18%, 15% and 18% in NOPL, TB and P, respectively.

This urban residential location S3, has its influence from various sources as it is located at the heart of the Guwahati city. Over past few years there is a tremendous increase in the number of vehicles and huge leap in construction activities all over Guwahati. It is surveyed that this number would double in every five years (http://pcbassam.org/). Also, there are two major city roads near S3 i.e., G.S road and R.G Baruah which are located at about 380 m and 450 m from the site. These busy roads are mostly occupied by two and three wheelers which emit 20-40% of fuel unburnt or partially burnt (Harish, 2012). Poor public transportation, uncontrolled and delayed bus stoppages in these commercial centres often create huge traffic congestion. Thus, the striking implications of huge traffic-related emissions were highlighted in the source profiles resolved for P (31%) mainly, along with NOPL (12%) and TB (19%).

Fig 4.13 shows the source profiles of rural area S4 along with percentage of species in a source. Predominant source was biomass burning in all the three regions i.e. NOPL (30%), TB (30%) and P (32%). Rapid urbanization and development of commercial centres in this

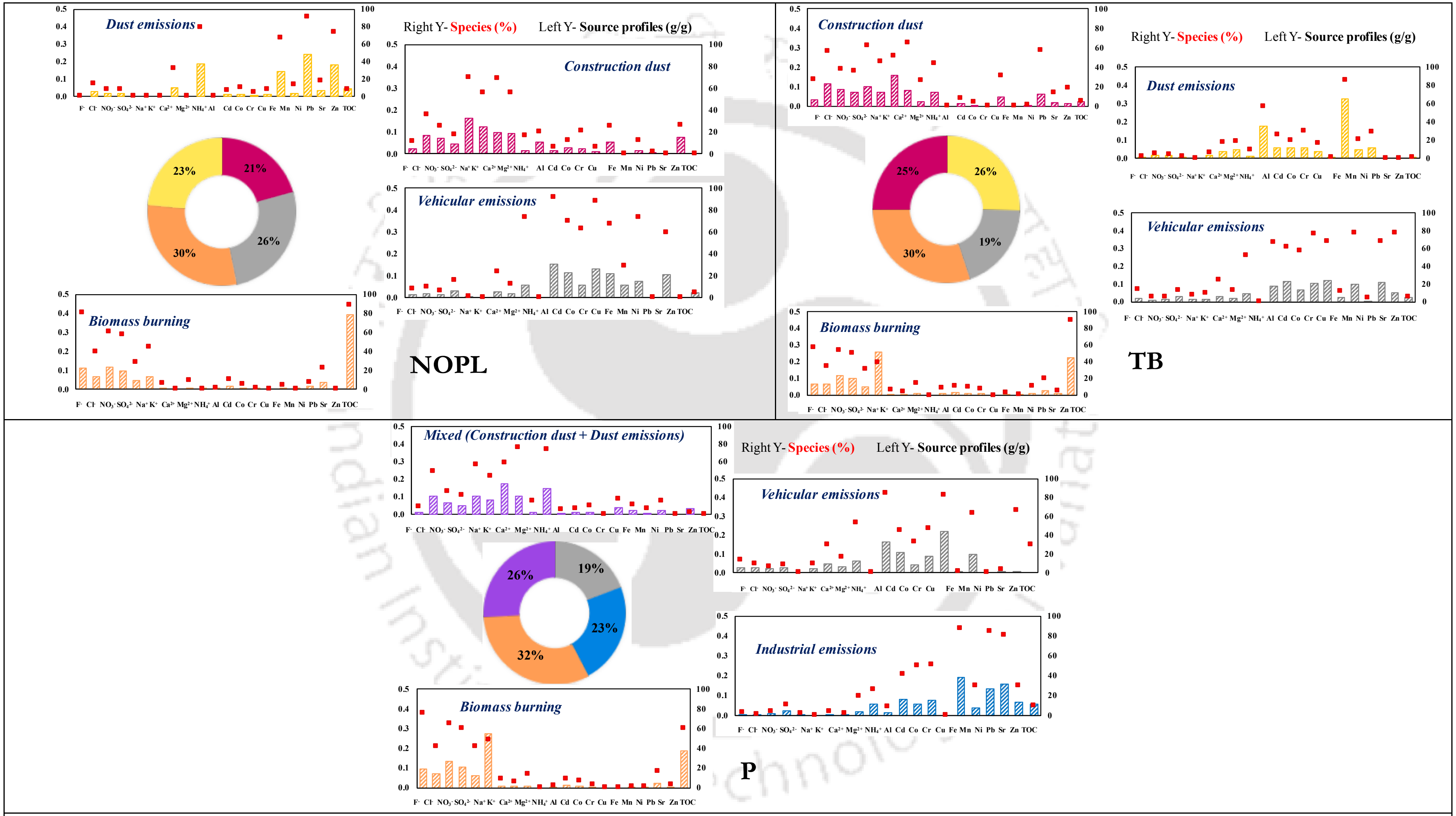


Fig 4.13 Source profiles resolved by EPA PMF v5.0 model for samples collected in S4 during the study period

region led to the accumulation of huge solid waste. And also the agricultural residues along with municipal solid waste has become the main source of biomass energy for domestic activities (Kataki et al., 2016). This might have resulted in the significant contribution of biomass burning in this area. The dust emissions and construction activities share a major proportion in NOPL (23% and 21%), TB (26% and 25%) and P (mixed-26%). The geography of Barpeta district varies from low-lying plains to highlands with slight hillocks sprawling over the south-west corridor of the district (http://barpeta.gov.in/geography.html). The major problem being faced by the Barpeta district is riverbank erosion caused by mighty Brahmaputra river and its tributaries like Beki river coupled with flood problem. The damage due to land loss ranged from 72.5 km^2/year in 1997 and 80 km^2/year between 2007-2008, indicates the severity of the issue in this area (Pal et al., 2013). So, the year wise construction of embankments takes place in this area to protect the area from flood havoc. Thus, the emissions from construction activities and dust emissions are inevitable and their effects were persistent in NOPL, TB and P regions. Also, the site has perfect amalgamation of town and village environment with no large-scale industries. This caused slight impact on the contribution of industrial emissions (23%) in P region alone. The site is located at about 660 m from the main market place and is well connected to major district road i.e. NH 31 which is about 25 m from the site. As the town is a district headquarter, there is a huge rush of people and the crowded public transport from all over the district during the office hours. This scenario was however reflected in the occurrence of vehicular emissions in NOPL (26%) and was decreased by 1.4 times in both TB and P regions.

Fig 4.14 shows the source profiles of suburban area, S5 along with percentage of species in a source.

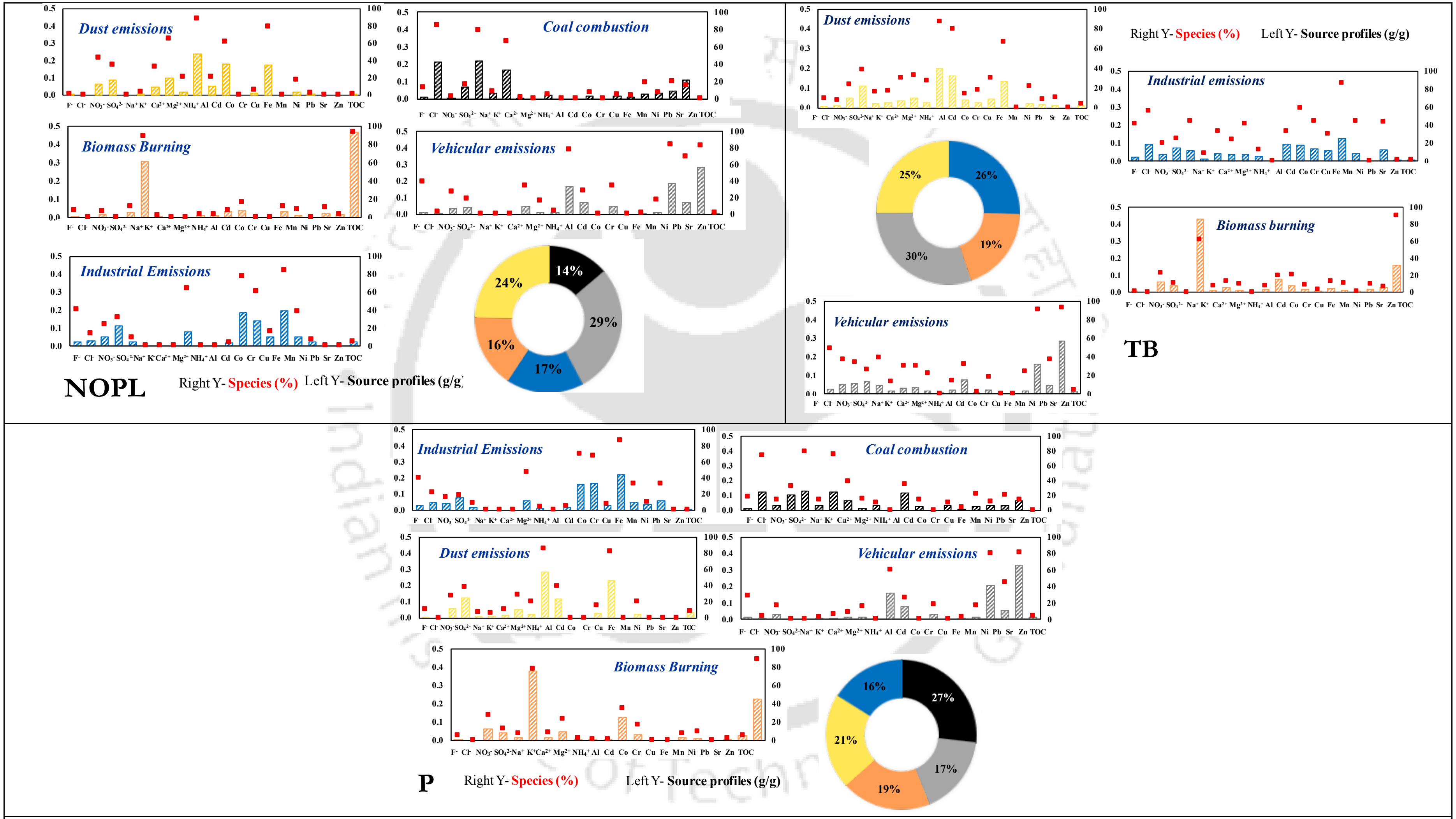


Fig 4.14 Source profiles resolved by EPA PMF v5.0 model for samples collected in S5 during the study period

Five different sources were identified in this location with four of them common in NOPL, TB and P regions. Jorhat is one of the oldest towns of Upper Assam, located on the eastern part of Brahmaputra valley. The considerable population growth led to the increase of number of vehicles like any other city, thereby burdening the current infrastructure to accommodate these vehicles. Traffic congestion during the peak hours is a very common sight in this area which has resulted in the findings of vehicular emissions in NOPL (29%) and TB (30%) while the reduced emissions by 1.8 times in P region. Also, the breadth of the roads in most of the places in Jorhat, remained smaller since past, allowing the passage for both slow-moving and motor transport. According to Jorhat municipal board, out of 39 km of total municipal road, 28 km is left unpaved with only 11 km being paved (http://jorhat.gov.in/). Such scenario in this suburban area would have resulted in the prevalence of dust emissions leading to 1/4th contributions in NOPL, TB and P regions. Industrial emissions were high in TB (26%) than NOPL (17%) and P (16%) regions. This could be due to rise of many small-scale industries in and around Jorhat town after the second world war (http://dcmsme.gov.in/dips/old_dipr.html).

Majority of the rural population in Jorhat depends on biomass resources such as crop residues, wood fuels and solid waste for various activities like cooking and heating purposes (Sarmah et al., 2002). Thus, the emissions from biomass burning were evident in NOPL (16%), TB (19%) and P (19%) regions. Finally, the coal combustion which is an important source in this region has its contribution mainly in NOPL (14%) and P (27%) with nil impact in TB region. 60 billion out of 861 billion tons of world's coal reserves, belongs to India. Of which, 1.597 billion tons originates from northeast India (Dutta et al., 2017). As the state is rich in natural resources such as coal, oil and natural gas, the open-cast coal mining activities in the upper Assam operated in five different collieries i.e. in Ledo, Baragolai, Tirap, Tikak and Tipong the influence of it is highly experienced in this area. As mentioned earlier, the area is surrounded by small-scale tea industries where coal is used as main energy source. Thus, this source is obvious in S5.

4.2.4 Comparison of source contributions in urban and rural sites

The comparison of overall source contributions resolved using PMF model in urban and rural areas have been compiled in Table 4.5. In general, three sources were common in all the sites: biomass burning, dust emissions and vehicular emissions.

In urban sites i.e. S1, S2 and S3, vehicular emissions were the predominant source with increased emissions in P than TB and NOPL. The release of ultrafine PM into the atmosphere due to vehicles in many urban areas was reported earlier (Morawska et al., 2008). The vicinity of the vehicular sources, increased traffic congestion and poor vehicle maintenance with low quality fuel has been reported by the earlier surveys conducted in this region (https://www.c-nes.org/1461/c-nes-report-on-air-pollution-released/). This could be the possible reason for the dominance of vehicular emissions in P region which can settle in the deeper sections of respiratory system. Secondary aerosols contributions were resolved only in urban areas i.e. S1 and S3. As it is understood, that in the urban environment the primary sources would be huge consumption fossil fuels in industries and transportation. These principal sources account for enormous emissions of SO_x and NO_x in urban areas, which are gaseous precursors of secondary aerosols (Venkataraman et al., 2018).

Due to the nearness to oil refinery plant, the relevant contributions of it was evident in S2 (18%) both in NOPL and TB. The pattern of dust emissions had a slight variation in the case of S2 with increased contribution in P than NOPL and TB. While in the case of S1 and S2, the variation was relatively constant with high contributions in NOPL and TB than P. The fact that these urban sites are developed as industrial areas with many shuttered factories, the industrial emissions have accounted for considerable share i.e. 17-28% in the deposition of PM in the human respiratory tract. The pollution due to year-round constructions activities in S1 was indicated in the contributions of 25% in TB and 22% in P regions.

Table 4.5 Overall source contributions (%) obtained using PMF model in all the sites

Site	Biomass burning	Coal mining	Construction dust	Dust emissions	Industrial emissions	Oil refinery	Secondary aerosols	Vehicular emissions
Nasopharyngeal (NOPL)								
S1	22	–	9	23	–	–	**35**	11
S2	**31**	–	–	19	12	18	–	20
S3	17	–	**23**	18	13	–	17	12
S4	**30**	–	21	23	–	–	–	26
S5	16	14	–	24	17	–	–	**29**
Tracheobronchial (TB)								
S1	12	–	25	25	–	–	–	**38**
S2	**27**	–	–	19	18	18	–	18
S3	**27**	–	–	15	18	–	21	19
S4	**30**	–	25	26	–	–	–	19
S5	19	–	–	25	26	–	–	**30**
Pulmonary (P)								
S1	7	–	22	17	15	–	–	**39**
S2	–	24	–	**28**	24	–	–	24
S3	20	–	–	17	16	–	18	**29**
S4	**32**	–	26		23	–	–	19
S5	19	**27**	–	21	16	–	–	17

In the rural sites, S4 and S5, much of the contributions stems from dust emissions and biomass burning, which is an obvious problem in rural areas due to the high proportion of unpaved dirt roads (Khan and Strand, 2018; Majra, 2011). Surprisingly, the vehicular emissions also showed significant contributions (17-30%) in these rural sites exceptionally in NOPL and TB regions. This can be understood due to the fact that during the last few decades, the intensified anthropogenic activities in rural areas have aggravated the air crisis (Xiao et al., 2016). Moreover, the coal mining and its subsequent activities can have an immediate impact on the air quality and this was noticed in the case of S5, where 27% and 14% in P and NOPL regions were reported.

4.2.5 Seasonal variation of source contributions of dominant sources in all the sites

The seasonal variation of predominant sources i.e. biomass burning, dust emissions and vehicular emissions in the all the five locations were given in Fig 4.15.

Northeastern region is covered by plenty of forestlands and the residues generated from them are the prime source of energy (Roy, 2013). Biomass burning in a state like Assam comprises of 50-65% of crop-burning and the rest as forest fires (Kataki et al., 2016). Forest survey of India (FSI) reported increasing number of forest fire incidents every year in this region, due to shifting of cultivation and other associated human activities (FSI, 2019). Further, the report stated that in 2018, these incidents have increased by a factor of 1.5 over the previous year. However, the contributions of biomass burning varies significantly at an urban-rural level along with seasons, such as open-cast burning in summer to heating purposes in winter (Chowdhury et al., 2012). Thus, for the present study the variation of biomass burning contributions, during winter and other seasons were compared as shown in Fig 4.15 (a). In NOPL and TB, the contributions of rural areas were 1.5-3.8 times higher in winter, while in the case of urban areas (S1 and S3) the emissions were reduced by 1.3-1.6 times in winter than the other seasons. However, the contributions in P region were slightly higher in other seasons by 1.1 times than winter in urban and rural areas. This phenomenon suggests that irrespective of household fuel usage, economic status or geographic location, the extent of pollution due to biomass burning was enormous and has equal consequences both in urban and rural scale. Such similar observations were reported earlier by numerous source apportionment studies in India as well in neighboring countries like China, across urban and rural environments (van Ruijven et al., 2011; Zhang et al., 2010).

Fig 4.15 (b) shows the variation of dust emissions in winter, pre-monsoon and monsoon.

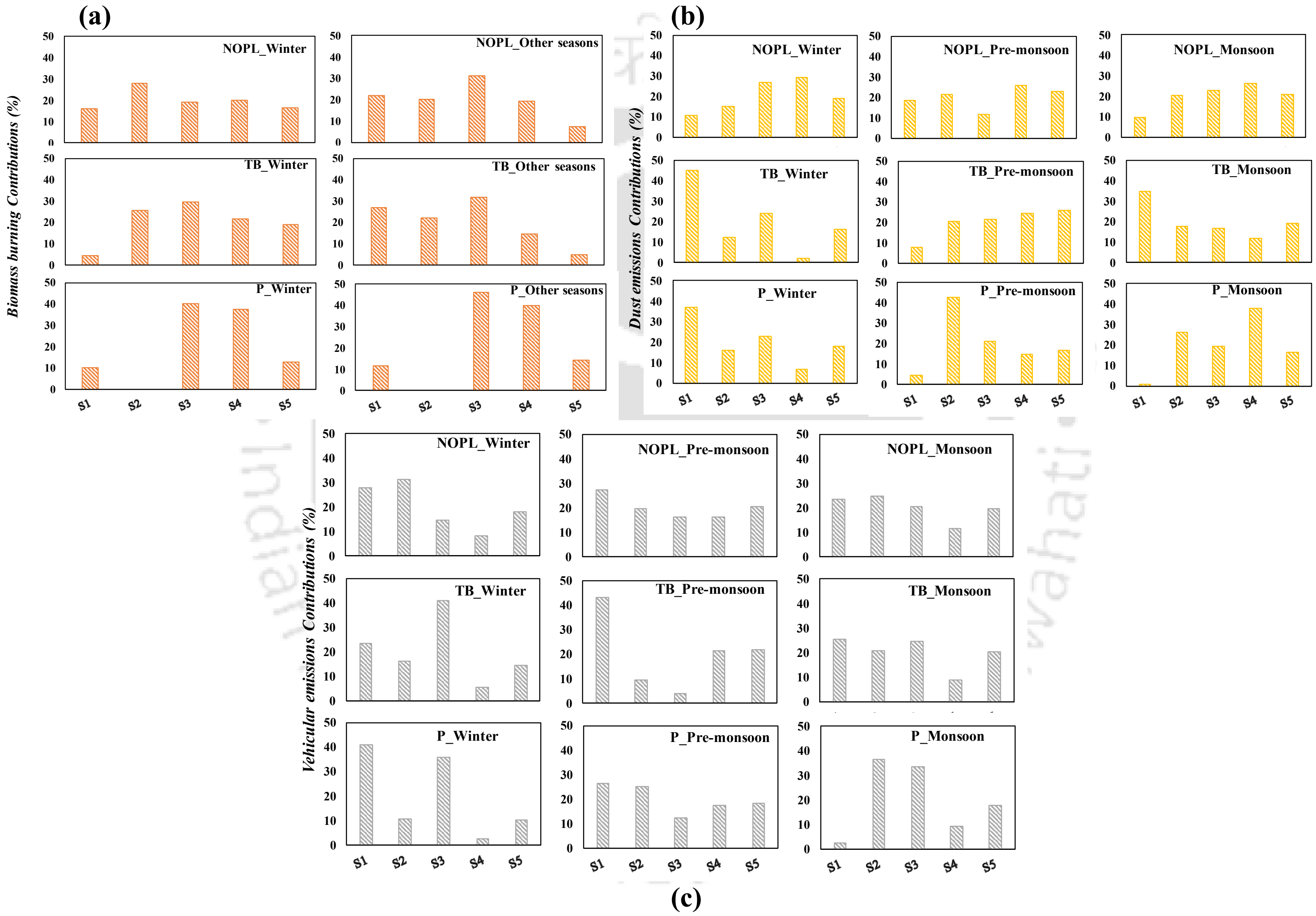


Fig 4.15 Seasonal variation of source contributions of (a) biomass burning, (b) dust emissions and (c) vehicular emissions in all the sites

In NOPL, the average contributions of dust emissions were high in winter than in pre-monsoon and monsoon in rural areas (24%) than in urban areas (17%). While in TB and P, the emissions were reduced by 1.16 and 1.2 times, respectively in rural areas than urban areas. This suggests that the dust emissions in rural areas are effecting NOPL region than TB and P which could be possibly due to the coarser particles of both wind-blown and mechanically generated. These coarse mode particles are generally high in winter than other seasons (Srimuruganandam and Nagendra, 2010).

The seasonal variation of contributions of vehicular emissions in NOPL, TB and P in all the sites were shown in Fig 4.15 (c). The average contributions of vehicular emissions in urban areas was higher in all the seasons than rural areas. A recent findings reported that 90% of urban pollution is due to vehicular emissions in evolving countries like India. (Shrivastava et al., 2018).

4.2.6 Episodic analysis at a selected location (S1)

In general, PM concentrations are high in winter and coincidence of Diwali festival during this period every year is a most noteworthy aspect. Many Indian studies reported the worsening of air quality in the aftermath of episodic pollution due to burning of fireworks and farm fires during Diwali. Therefore, the air pollution analysis around this period which is usually celebrated for three consecutive days, may yield some useful and interesting conclusions.

In the present study, Nov'7th, 2018 was Diwali and the episodic analysis for a period of six days in one of the sites i.e. S1, was conducted to understand the effect of burning fireworks on the human respiratory system. For this purpose, the CMB approach was used to identify the source contributions using the measured speciated data matrix during Diwali period and source profiles utilized from PMF analysis (Fig 4.16). The source profile of fireworks was derived from literature of Indian studies (Tian et al., 2014).

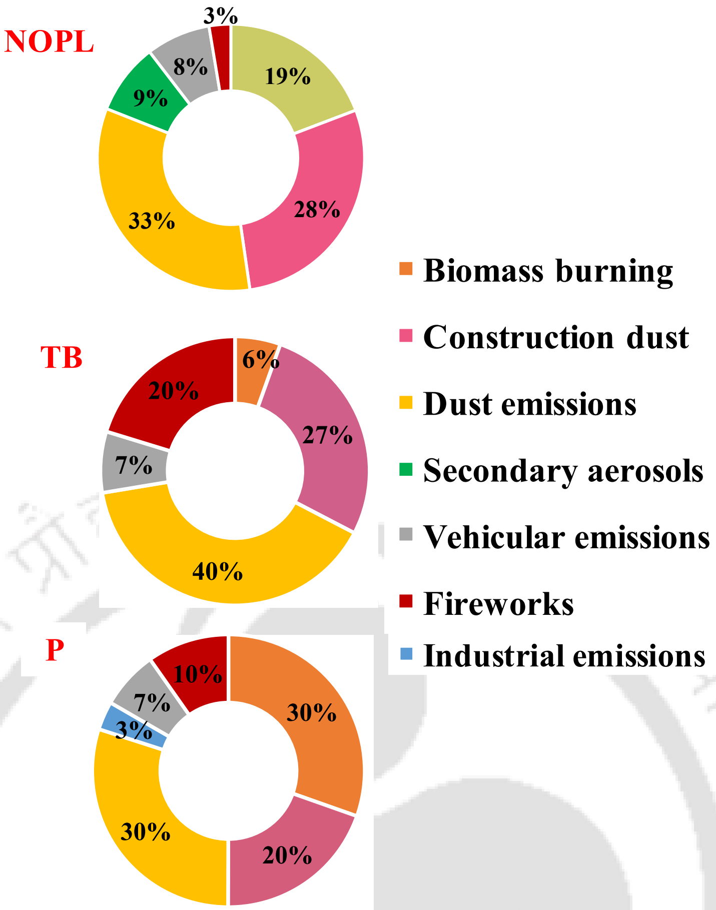


Fig 4.16 Source contributions during Diwali and their variation of regional deposition

As shown in Fig 4.16, there has been substantial variation in the distribution of source contributions during Diwali in NOPL, TB and P regions. While, the burning of huge fireworks exhausts within few hours, but the degradation of air quality is tremendous and effects on human health are enormous. In the past, several studies indicated the interrelation between increase in PM and acute health effects caused due to burning of fireworks. To find the risk due to inhalation of metals produced during this episode, a short-term study along with health based hospital survey was conducted during Diwali'2015 and the following facts were observed:

Panel (a) in Fig 4.17 indicates that the number of patients suffering from headache, fatigue, irritation in eyes, coughing, sneezing and sinusitis, increased by 3.8, 3, 3.3, 3.3, 3.5 and

6.5 times, respectively on Diwali days compared to non-Diwali days. Similar findings have been found in a post-Diwali morbidity survey conducted at Delhi (Sharma et al., 2015). Also, five major metals (Cd, Co, Fe, Zn and Ni) were used to assess the possible non-carcinogenic human health risks due to this pollution episode. Two scenarios were taken into consideration. In scenario I, cumulative Hazard Index (HI) due to metals (Cd, Co, Fe, Zn and Ni) for an annual period (350 days) was estimated using average concentrations during non-Diwali days. 350 days is based on U.S EPA guidelines (USEPA, 2004) which describes this value as the most frequent exposure that is reasonably expected at a site with two weeks of vacation. The degree of over or under estimation is considered to be negligible.

Scenario II involved two parts (a) and (b). In II (a), HI was calculated for all the metals for 347 days, excluding Diwali period, by utilizing concentrations as in scenario I. In II (b), HI for the Diwali period was calculated. Summation of II (a) and (b) was compared with HI from Scenario I to get the excess risk due to Diwali.

Non-carcinogenic health risk to an individual due to exposure to all metals '*j*' is given by Eq 13 (U.S.DOE, 1999):

$$HI = \sum_{j=1}^{k} \frac{M_j \times EF \times ED}{AT \times RfC_j} \qquad \textbf{(13)}$$

where, RfC is the inhalation chronic reference concentration (mg/m^3) obtained from: The Risk Assessment Information System (RAIS) (Sharma et al., 2015), M is concentration of a metal (mg/m^3), EF is exposure frequency (days/year), ED is Exposure duration (years), and AT indicates averaging time [age (years) × 365 (days/years)].

Using data collected from patients, and concentrations measured during the analysis, the

Reproduced with permission from: Garaga R and Kota SH. Characterization of PM$_{10}$ and Impact on Human Health During the Annual Festival of Lights (Diwali). Journal of Health and Pollution 2018; 8 (20).

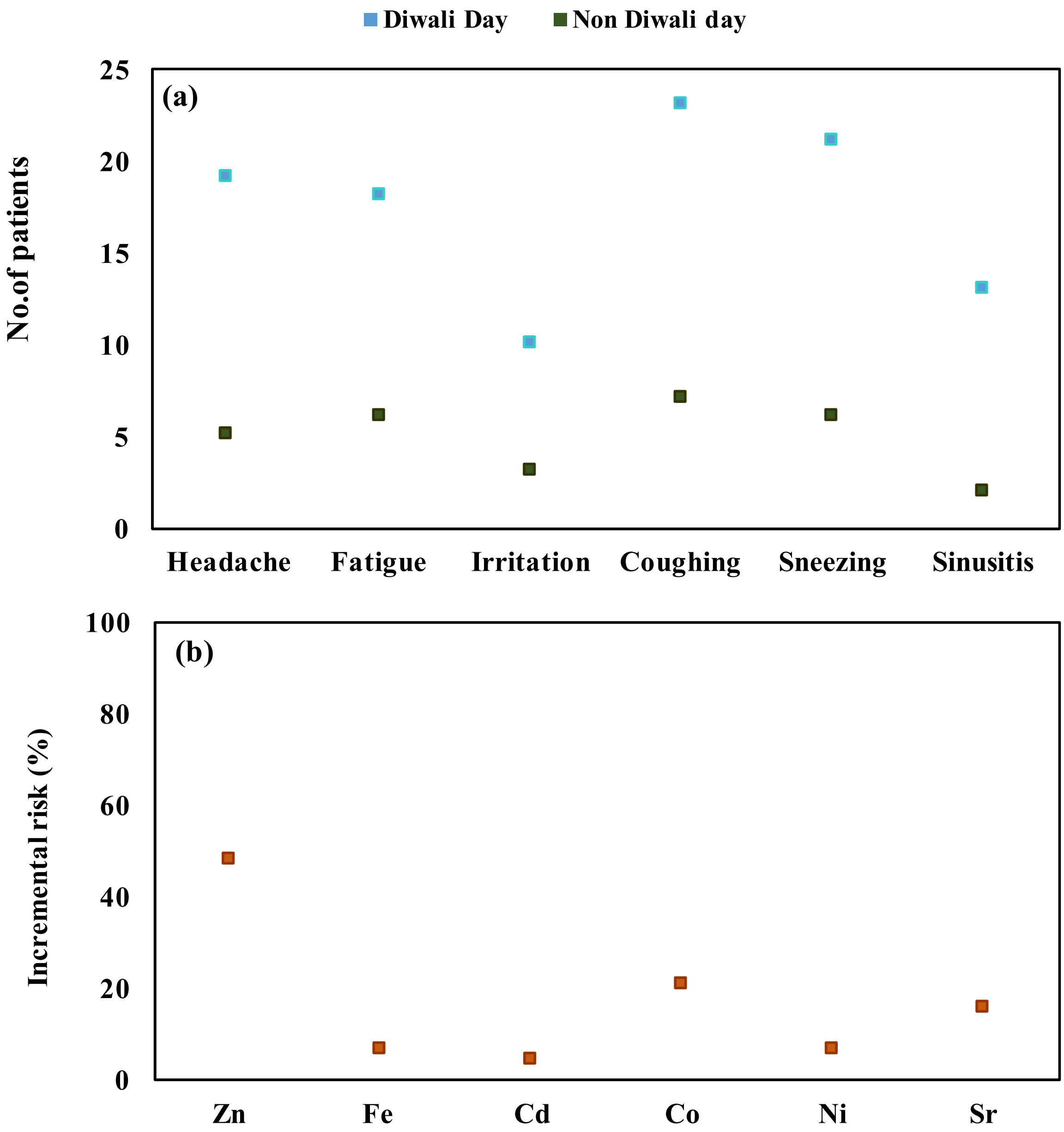


Fig 4.17 (a) Number of patients suffering from headache, fatigue, irritation, coughing, sneezing and sinusitis during Diwali and Non-Diwali days, and (b) Incremental risk (%) due to Diwali from metals

percentage incremental risk (i.e. the percentage increase between Diwali and non Diwali days) due to each metal is estimated using Eq 14 and shown in panel (b) of Fig 4.17.

$$\% \text{ increase} = (New\ value - Initial\ value) / (Initial\ value) \times 100 \quad \mathbf{(14)}$$

Estimated Incremental risk was highest for Zn (1.013), followed by Co (1.005), Sr (1.004) and the remaining elements: Ni (1.0017), Fe (1.0016) and Cd (1.0011). Results indicated that cumulative HI for Scenario II which includes Diwali period showed a risk level increase of ~ 0.5% in health effects due to chronic lifetime exposure to metals alone. It should be noted that this risk level rise was only due to metals, and in ambient settings the risk level will be higher due to chronic exposure to many other pollutants like SO_2, NO_x and EC, etc., released by fireworks.

This health study in Diwali'2015 showed the possible impact of fireworks on human health. Also, the present study strongly suggested that these effect were localized to respiratory system by depositing in TB and P regions of human respiratory tract.

4.2.7 Comparison of sources between wet and dry depositions at a selected location (S1)

Using the PMF method, sources of PM chemical composition were apportioned in the wet and dry depositions. The relative source contributions were compared since, similar source profiles were obtained between two different sampling types. In order to compare the sources identified in wet and dry depositions, urban site S1 was selected since, the sampling location of wet deposition was close to S1, with an aerial distance of 8 Km.

As shown in Fig 4.18, dust emissions contributed to 29% of the total source contributions in the dry deposition samples at S1, followed by secondary aerosols (23%), construction dust (20%), vehicular emissions (19%) and biomass burning (9%). In total, these factors accounted for 53% of PM in the wet deposition samples, with 26% of dust emissions, 18% of vehicular emissions and 9% of secondary aerosols. The remaining 47% of the contributions were due to industrial emissions (19%) and marine source (28%). The common and dominant sources were dust emissions (26% in wet and 29% in dry) and vehicular emissions (18% in wet and 19% in dry). The contribution of the dust emissions and secondary aerosols in dry deposition samples was 52% on average (29% + 23%) which apportioned half of the total source contributions. While in the case of wet deposition, the contribution of dust emissions, secondary aerosols and marine source related factors was 63% which was far higher than industrial and vehicular emissions of 37% on average. This indicates the effective washout of coarser particles due to wet scavenging process (Huston et al., 2012). Therefore, the present study indicated the influence of anthropogenic sources in case of both wet and dry depositions.

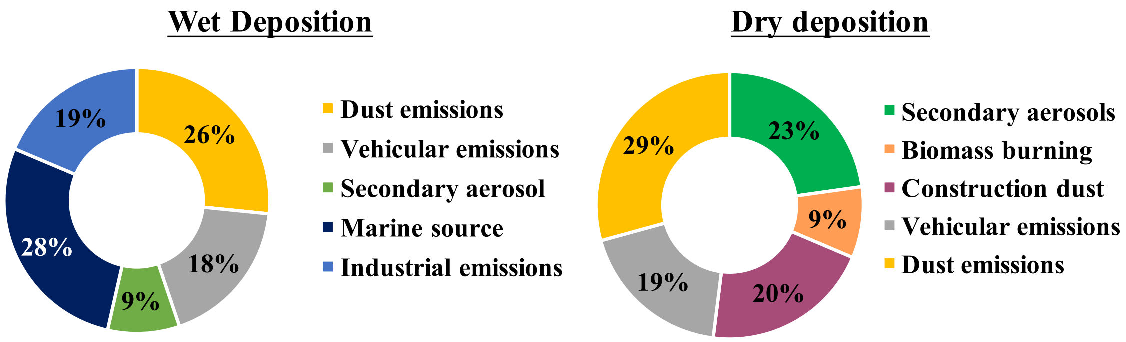


Fig 4.18 Comparison between the sources of wet and dry depositions in site S1

4.2.8 Summary

Size resolved PM was monitored during Jan’2018-Dec’2018 in five locations of Assam and following observations were recorded: Concentrations of PM_{10} and $PM_{2.5}$ in most of the sites have exceeded the prescribed limits of air quality. Regional deposition in P was high in S1 and S3 when compared with others. Vehicular emissions were dominant in both S1 and S3 in P while biomass burning being dominant in S3 which could be the reason for maximum deposition in P. Dust emissions was predominant source in individual masses of PM, while size resolved PM revealed vehicular emissions being dominant in ultrafine scale. Clear distinction of source types in urban (S1, S2 and S3) and rural (S4 and S5) areas was observed, where dust emissions and biomass burning were high in rural and vehicular emissions being maximum in urban areas. The impact of oil refinery is confined to local contribution at the urban site (S3), having negligible influence on other two urban sites (S1 and S2). Also, the coal mining activities were identified to have its maximum contribution at the rural site (S5), strictly demanding the regulation strategies to be implemented. Episodic analysis revealed more significant influence of firework emissions in TB and P than in NOPL regions of human respiratory tract indicating the necessity of regulated burning of fireworks in regions with high population density.

CHAPTER V– CONCLUDING REMARKS

5.1 SUMMARY OF THE THESIS

The seasonal variations in characteristics of wet and dry depositions of PM is determined in Assam, a northeastern state of India. Due to the heterogeneous nature of PM, its variations depend largely on natural geographical factors such as vegetation, topography and also prevailing meteorological conditions. Therefore, the determination of PM differs from region to region and thus, demands the site/region specific analysis.

Therefore, the main objective of present study is to investigate the sources of wet and dry depositions of PM in Assam. Rainwater analysis for an urban area was conducted over a period of one year i.e. June’2016-June’2017. Size resolved PM was collected during Jan’2018 to Dec’2018 in five locations (i.e. three urban and two rural areas) of Assam. Detailed chemical characterizations of both wet and dry depositions using advanced laboratory methods were carried out. In addition, the deposition of size resolved PM in human respiratory tract was evaluated. Also, the isotope analysis for the rainwater samples collected was carried out to locate the moisture source of rain droplet. Results obtained from chemical characterization were further utilized to identify the sources and their contributions using the receptor oriented method. Critical observations and corresponding conclusions drawn are presented in this chapter along with the limitations and extendable future scope of the present study.

5.2 MAJOR CONCLUSIONS

Major conclusions drawn from the current study are presented below, based on the different aspects investigated.

- Wet deposition analysis reported very low pH with high SO_4^{2-} and NO_3^- concentrations which contributed to the occurrence of (76%) acid rain events throughout the year in this region.
- While in many other Indian cities where crustal sources played a major role in buffering the acidity of rainwater, poor neutralizing capacity of these species was observed in the northeastern region.
- Mass concentrations of PM_{10} and $PM_{2.5}$ monitored in all five different locations of Assam, exceeded the prescribed limits of National Ambient Air Quality Standard (NAAQS).
- Common sources for both wet and dry depositions of PM identified during this study were dust emissions (wet (26%), dry (29%)), vehicular emissions (wet (18%), dry (19%)) and secondary aerosols (wet (9%), dry (23%)).
- Sources of wet deposition have their origin from both local and regional sources such as industrial emissions (28%) and dust emissions (25%) during non-monsoon and marine source (40%) during monsoon.
- Probable sources responsible for the deposition of PM in different sections of human respiratory tract are predominantly: vehicular emissions (high in P in all the seasons, where 25% in urban areas where 13% in rural areas), dust emissions (high in rural areas (24%) during winter in NOPL, while high in urban areas during pre-monsoon in TB (27%) and P (30%)) and biomass burning (high in winter in rural areas by 1.5-3.8 times than urban areas, in NOPL and TB regions).

- The present study based on both wet and dry depositions indicated that this region of India is highly prone to air quality threat thereby requiring immediate control measures and harmonization of trans-boundary policies.

5.3 LIMITATIONS AND FUTURE SCOPE

It is obvious that any research always holds certain assumptions and limiting conditions. Therefore, the outcome presented in this thesis must be applied judiciously considering the below mentioned limitations. Any study is indefinite and always poses scope for further research. Following are some of the identified extendable research ideas from this thesis

- Future investigation on acidification of rainwater in other urban areas including rural areas of this region, is further recommended to find its influence on the ecosystems and human health.
- Comparision of results with different receptor models such as CMB, UNMIX and PMF would be helpful to interpret more significantly.
- An overall approach to reduce the air pollution in any region should consider regional strategies along with local strategies to improve the air quality for better environment and health.
- Apart from recording the database of PM mass concentrations, the continuous monitoring of PM speciation and its association with epidemiological studies would gain understanding in designing the mitigation strategies for air quality development.
- Developing the emission inventory in this region followed by regional air quality modeling would aid in estimating contributions of local and non-local sources to PM in this region.

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ANNEXURE A

Detailed description of each sampling location along with wind rose plots

S1 (Jalukbari Site)

Jalukbari is located at the periphery of western part of Guwahati city. Jalukbari is the conjuncture point of four major roads of Assam. So, there is always a huge rush of automobiles from all over Assam at Jalukbari. Besides there is huge construction work, carried over by NHAI. The site location is situated at about 400 m from the National Highway 37. Moreover, there are two industrial areas nearby Jalukbari. Amingaon industrial area is 3.5 km to the north of Jalukbari site. Boragaon industrial area is 4 km to the south of Jalukbari site.

S2 (Noonmati Site)

Noonmati is located on the east side of the heart of the Guwahati city. Noonmati boast of having Petroleum Refinery, Indian Oil Limited Refinery or better known as Guwahati Refinery. Besides the refinery there are also vehicular emissions. The site location is about 250m from the refinery. Petrochemical plant is associated with raised levels of Ni, V and Zn.

S3 (ABC Tarun Nagar Site)

ABC Tarun Nagar is located at the heart of the Guwahati city. Over past few years there is a huge leap in construction activity and automobiles all over Guwahati. There are two major city roads near the site location. G.S Road is located at about 380m from the site location and R.G Baruah Road is located at about 450m from the site location. There is always a huge traffic rush in both the roads.

S4 (Barpeta Site)

Barpeta is a town in lower Assam which is also a district headquarter of Barpeta District. It is located about 90 km north-west of Guwahati. As it is a district headquarter, there is a huge rush of people from all over the district during the office hours. There are no large scale industries as such in Barpeta town which releases pollutant in the atmosphere. Vehicular traffic is one of the main source of air pollutant. The site is located at about 660 m from the main market place. The instrument was placed at 25m from the major district road which connects NH 31. The site has perfect amalgamation of town and village environment. It is surrounded by wetlands on two sides.

S5 (Jorhat Site)

Jorhat is located at far East end of Assam. It is a major coal mining place in Assam.

ANNEXURE B

The recovery of the elements using urban particulate matter by National Institute of Standard and Technology (NIST), the Standard Reference Material (SRM) 1648a to estimate the reliability of the analytical procedures. Following Table A.1 shows the detection limits and recovery rates of the elements.

Table B.1 Detection limits (ppb), and the recovery rate (%) obtained using SRM 1648a

Element	Detection limit (ppb)	Measured value (ppb)	Certified value (ppb)	Recovery (%)
Al	0.3	29.36	31.04	94.6
Cd	0.3	29.82	35.1	85
Co	0.1	25.3	25.36	99.7
Cr	0.8	16.34	18.63	88
Cu	0.6	18.7	20.5	91.2
Fe	0.4	84.47	91.2	92.6
Mn	0.5	33.04	37.66	87.7
Ni	0.7	58.1	58.8	98.8
Pb	0.6	18.47	18.5	98.8
Sr	0.2	2.22	2.35	94.5
Zn	0.2	67.46	72.48	93.1

ANNEXURE C

The brief explanation of calculation of average deposition fraction in human respiratory tract for each size range has been explained below.

$$\textbf{NOPL}\ (y) = 0.31476+0.70466\,x+0.34162\,x^2-0.19944\,x^3-0.19381\,x^4-0.04381\,x^5 \tag{C.1}$$

$$\textbf{TB}\ (y) = 0.02326+0.04405\,x+0.02369\,x^2-0.04062\,x^3-0.00127\,x^4 \tag{C.2}$$

$$\textbf{P}\ (y) = 0.15877+0.20228\,x-0.00114\,x^2-0.62572\,x^3-0.06118\,x^4+0.26125\,x^5+0.0778\,x^6 \tag{C.3}$$

where, y is deposition fraction for three regions i.e. NOPL, TB and P calculated using the respective equations and x is aerodynamic diameter

Table C.1 Calculation of average deposition fraction for NOPL, TB and P regions in each size range

x	**0.01**	**0.4**	**0.7**	**1.1**	**2.1**	**3.3**	**4.7**	**5.8**	**9**	**10**
LOG(Size)	-2.00000	-0.39794	-0.15490	0.04139	0.32222	0.51851	0.67210	0.76343	0.95424	1.00000
NOPL_ ∫ydx	-0.15587	-0.07753	-0.04075	0.01364	0.14113	0.26861	0.38911	0.46788	0.64260	0.68504
Average deposition fraction	**0.04890**	**0.15133**	**0.27709**	**0.45399**	**0.64942**	**0.78460**	**0.86241**	**0.91570**	**0.92740**	
TB_∫ydx	-0.17595	-0.00652	-0.00311	0.00100	0.00994	0.01834	0.02587	0.03059	0.04049	0.04277
Average deposition fraction	**0.10576**	**0.01402**	**0.02094**	**0.03181**	**0.04281**	**0.04905**	**0.05168**	**0.05188**	**0.04985**	
P_∫ydx	-0.65723	-0.05079	-0.02225	0.00674	0.05997	0.09866	0.12338	0.13398	0.14477	0.14552
Average deposition fraction	**0.37854**	**0.11740**	**0.14773**	**0.18953**	**0.19708**	**0.16101**	**0.11598**	**0.05654**	**0.01646**	

Concentration of regional deposition of PM (µg/m^3) = (PM concentration in each size range (µg/m^3)) x (Average Deposition fraction in each size range)

Fractional deposition of PM = (Average deposition in each region) / (Sum of all concentrations of deposition of PM)

ANNEXURE D

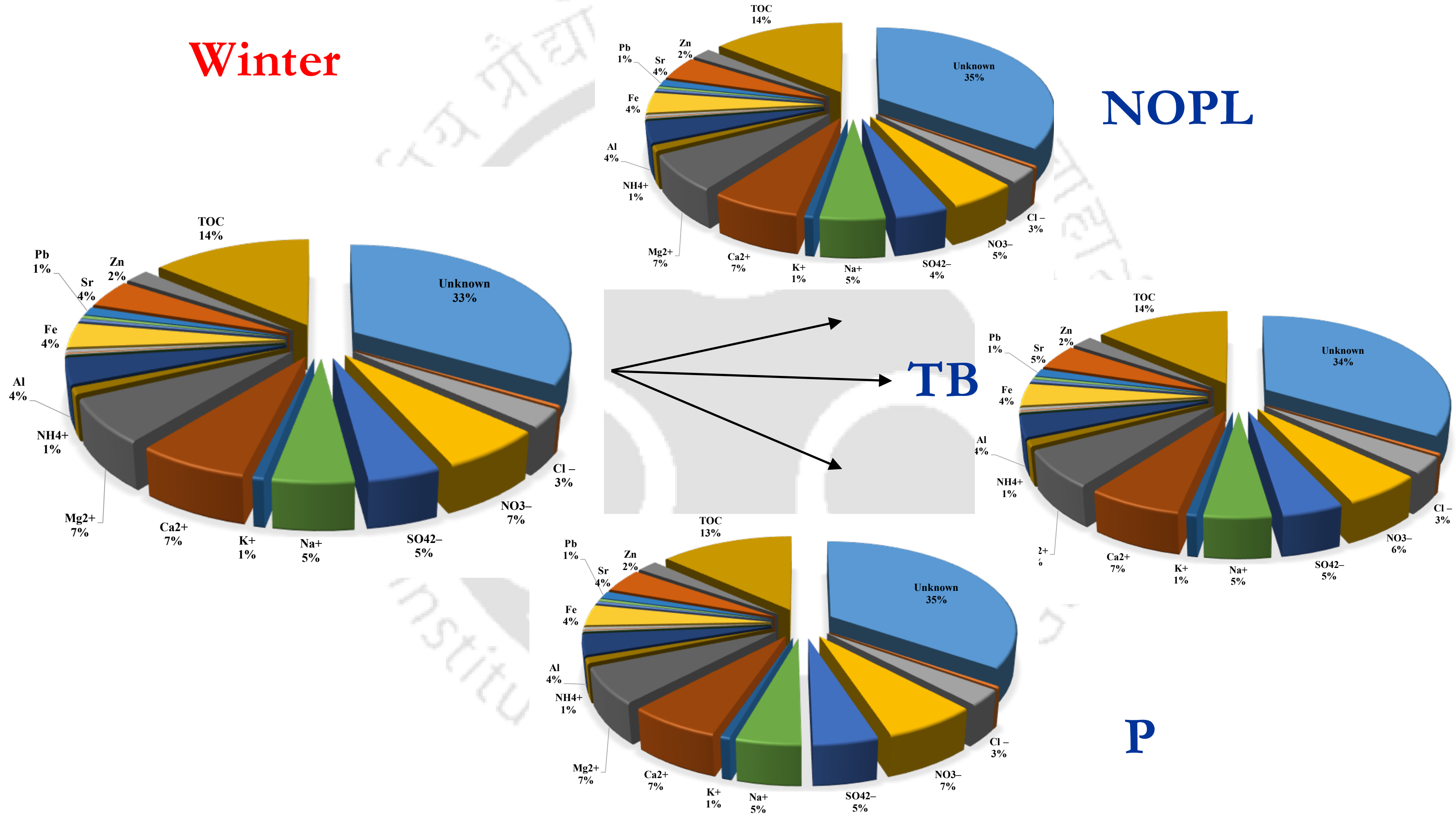

Fig D.1 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S1 during the study period

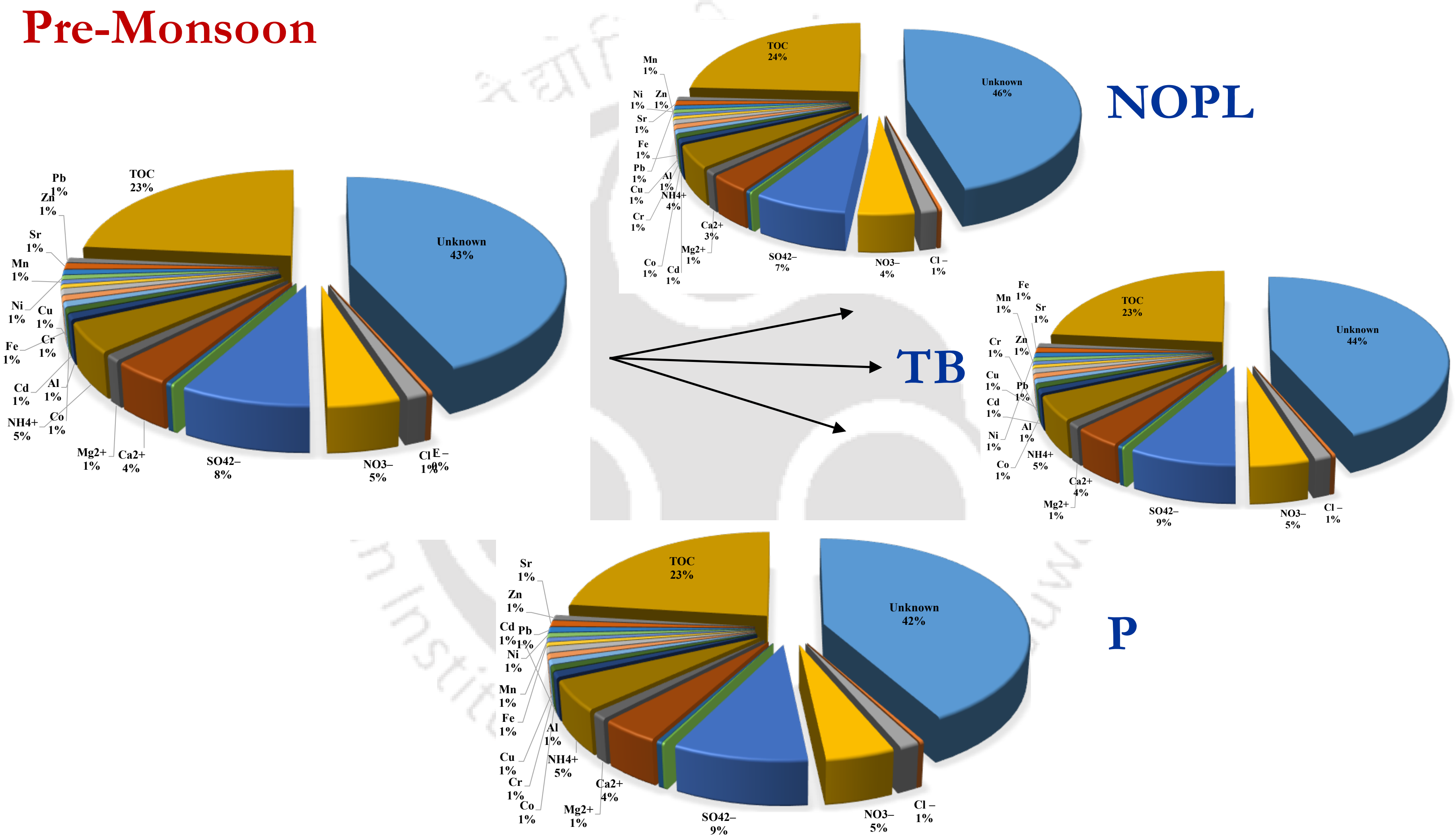

Fig D.2 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S1 during the study period

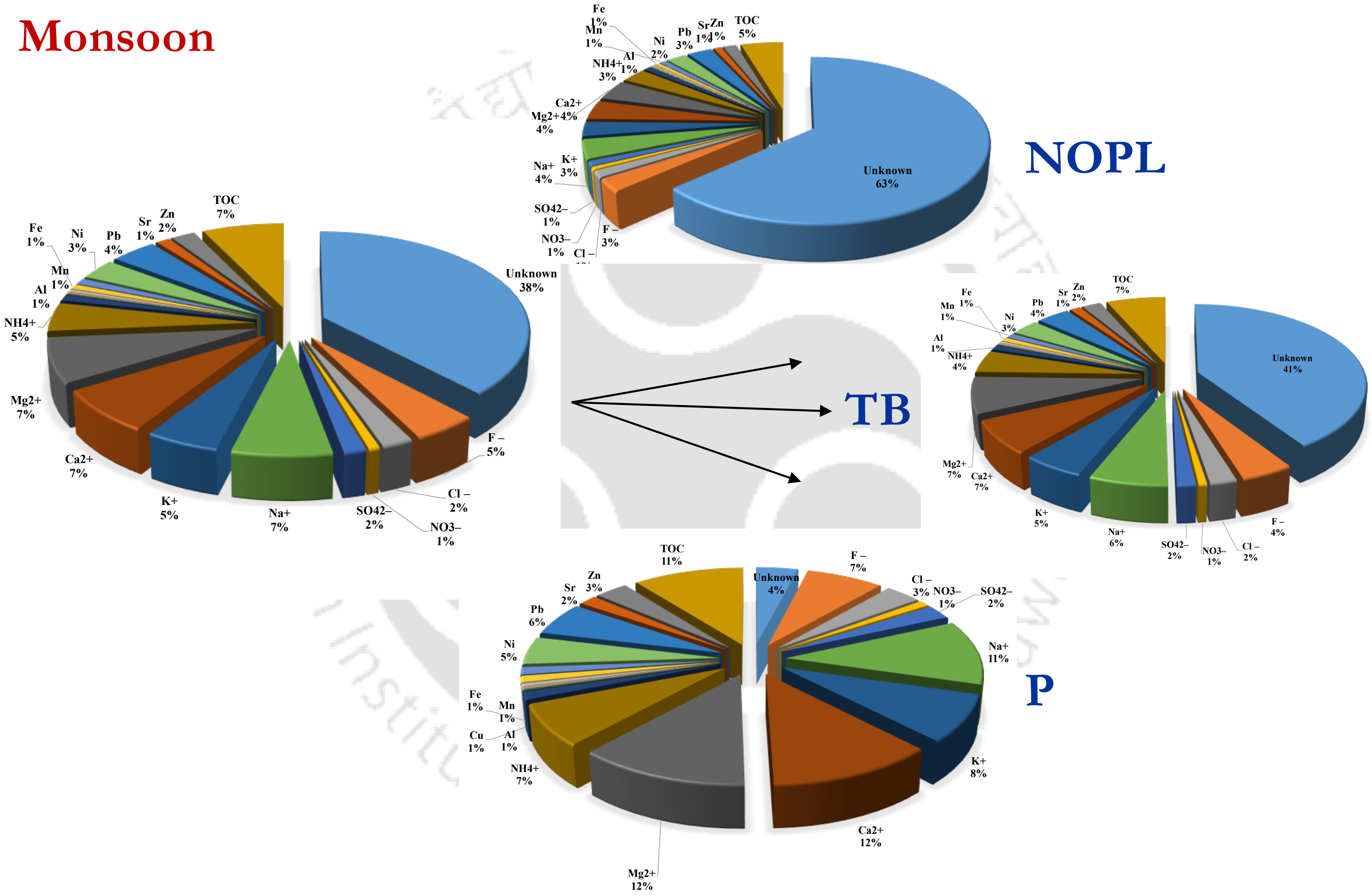

Fig D.3 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S1 during the study period

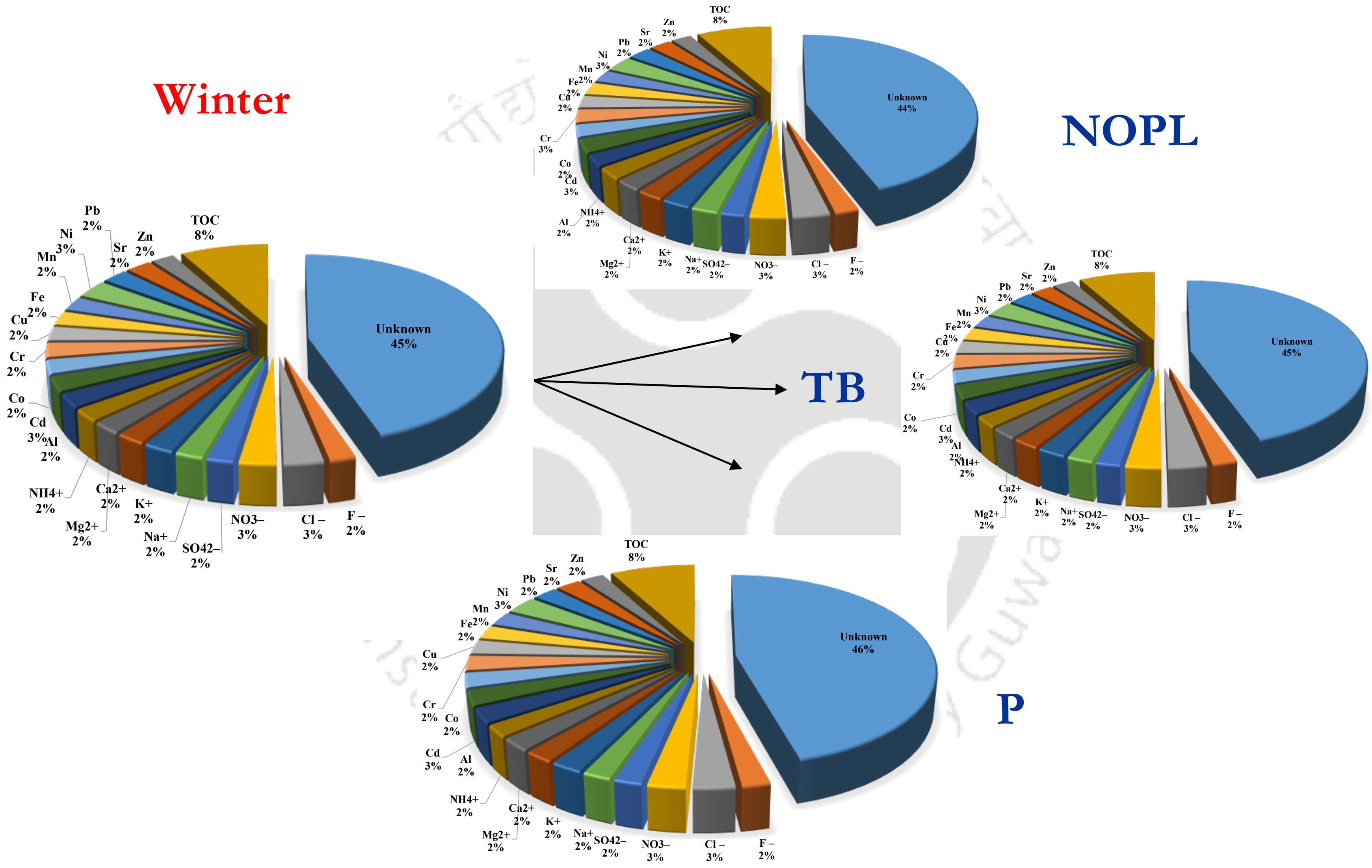

Fig D.4 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S2 during the study period

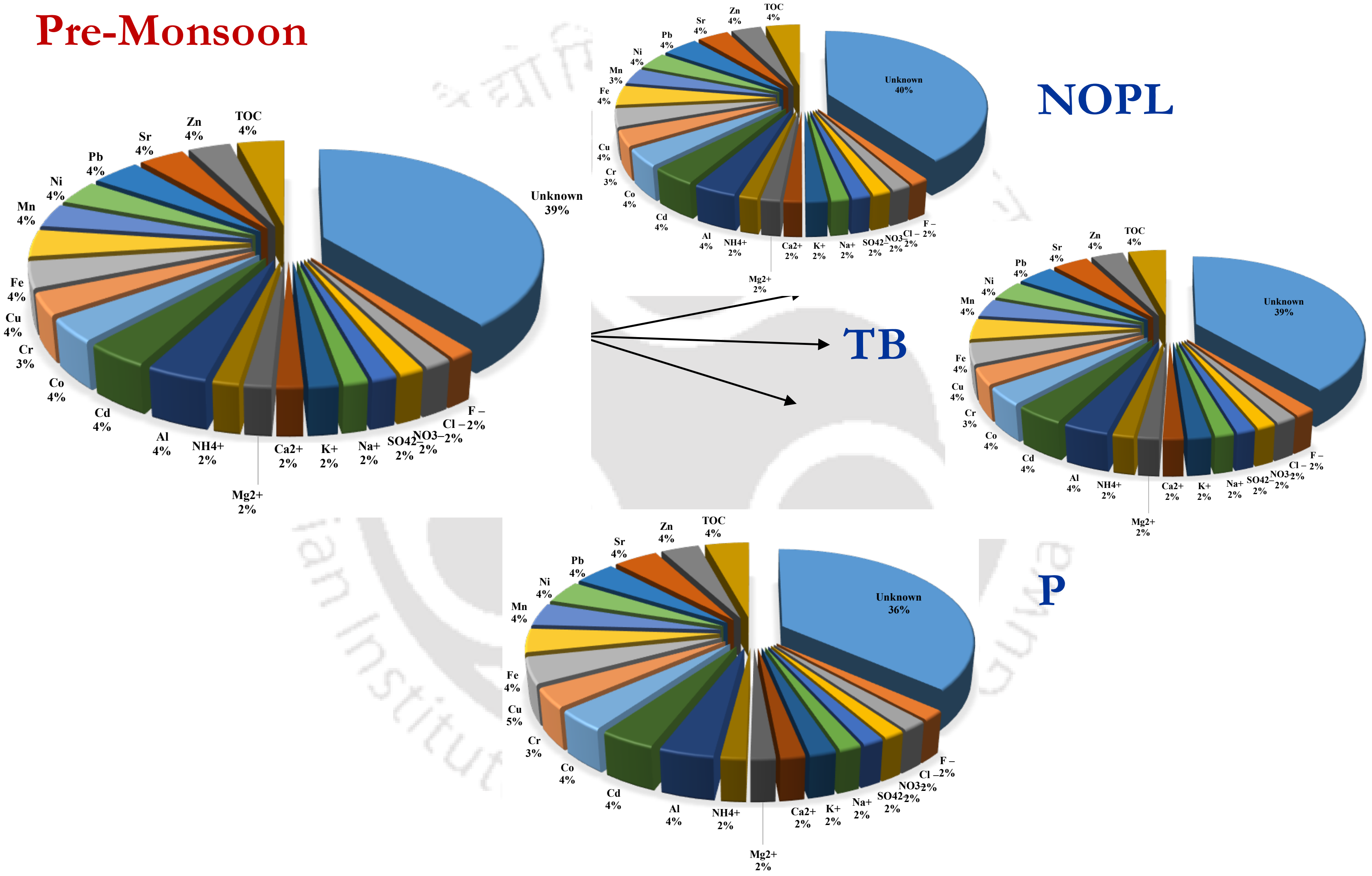

Fig D.5 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S2 during the study period

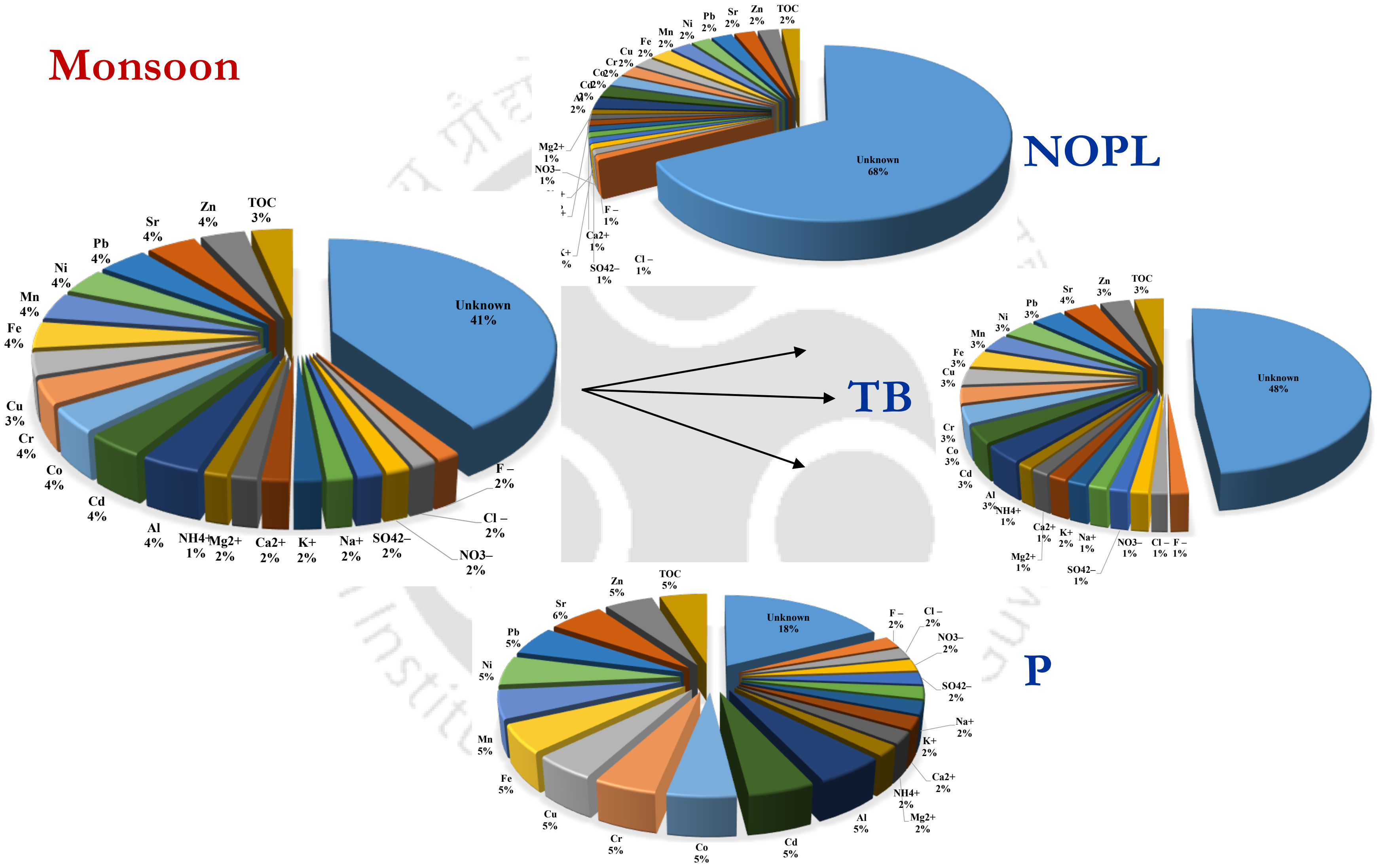

Fig D.6 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S2 during the study period

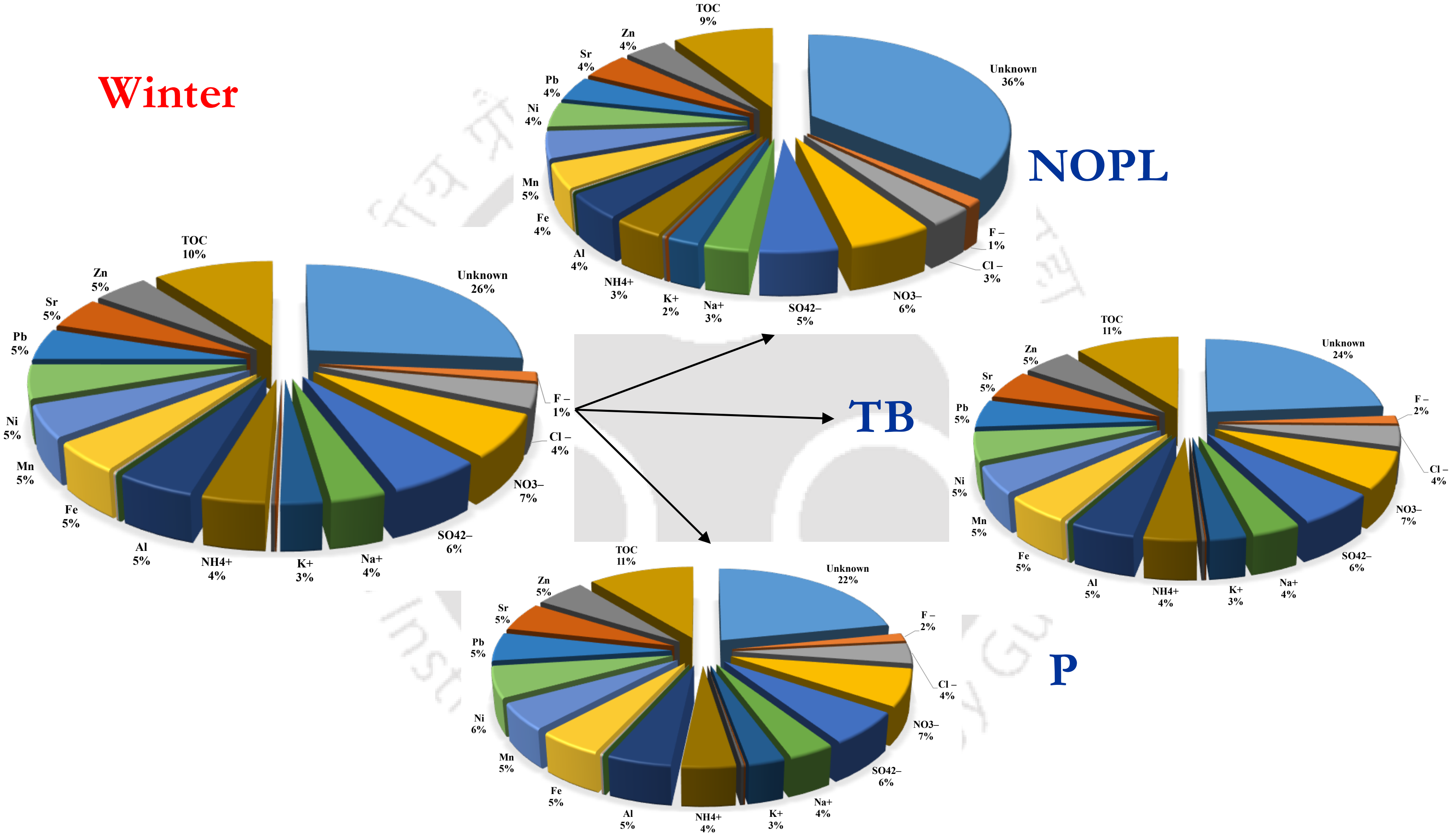

Fig D.7 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S3 during the study period

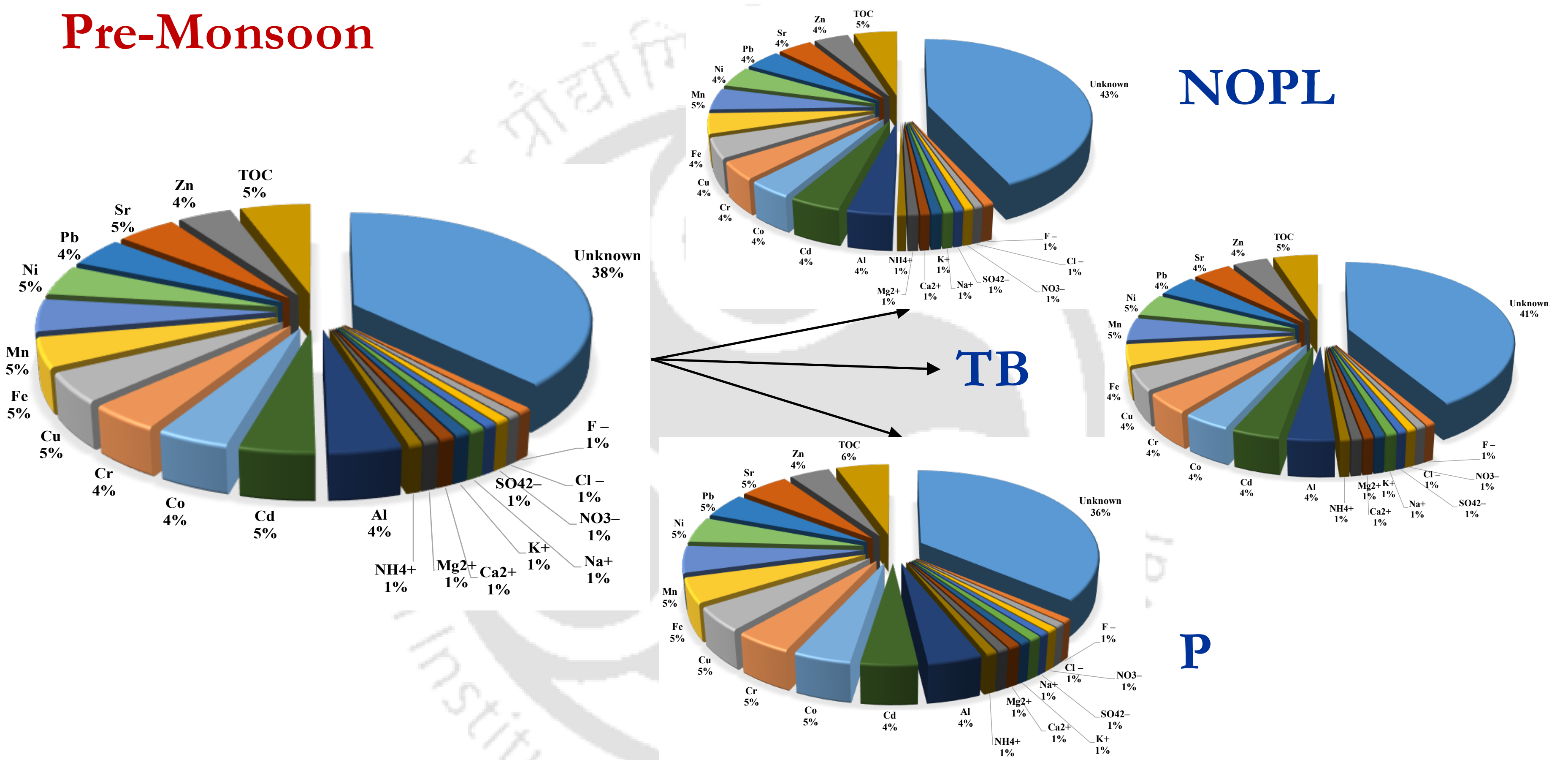

Fig D.8 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S3 during the study period

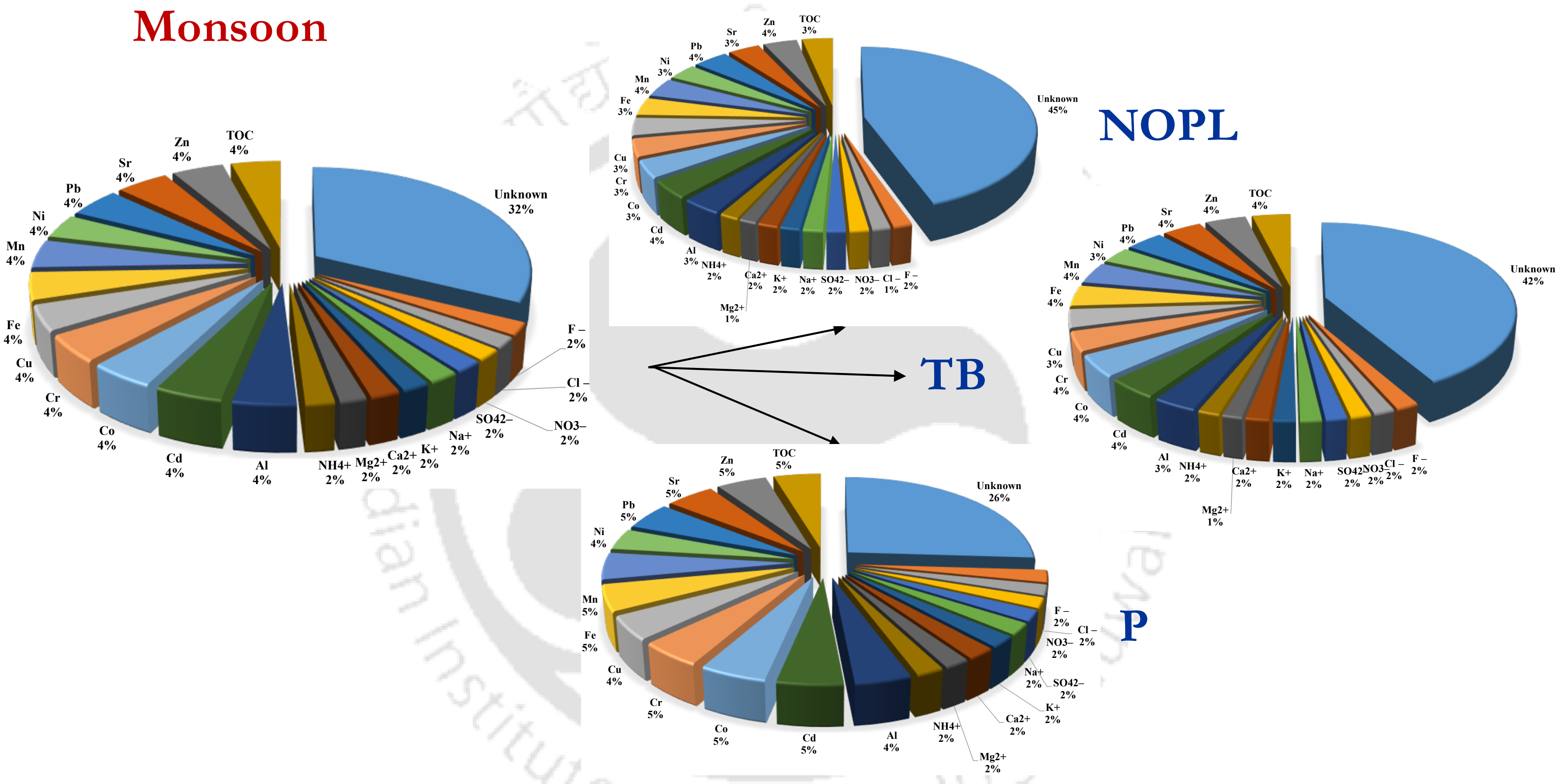

Fig D.9 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S3 during the study period

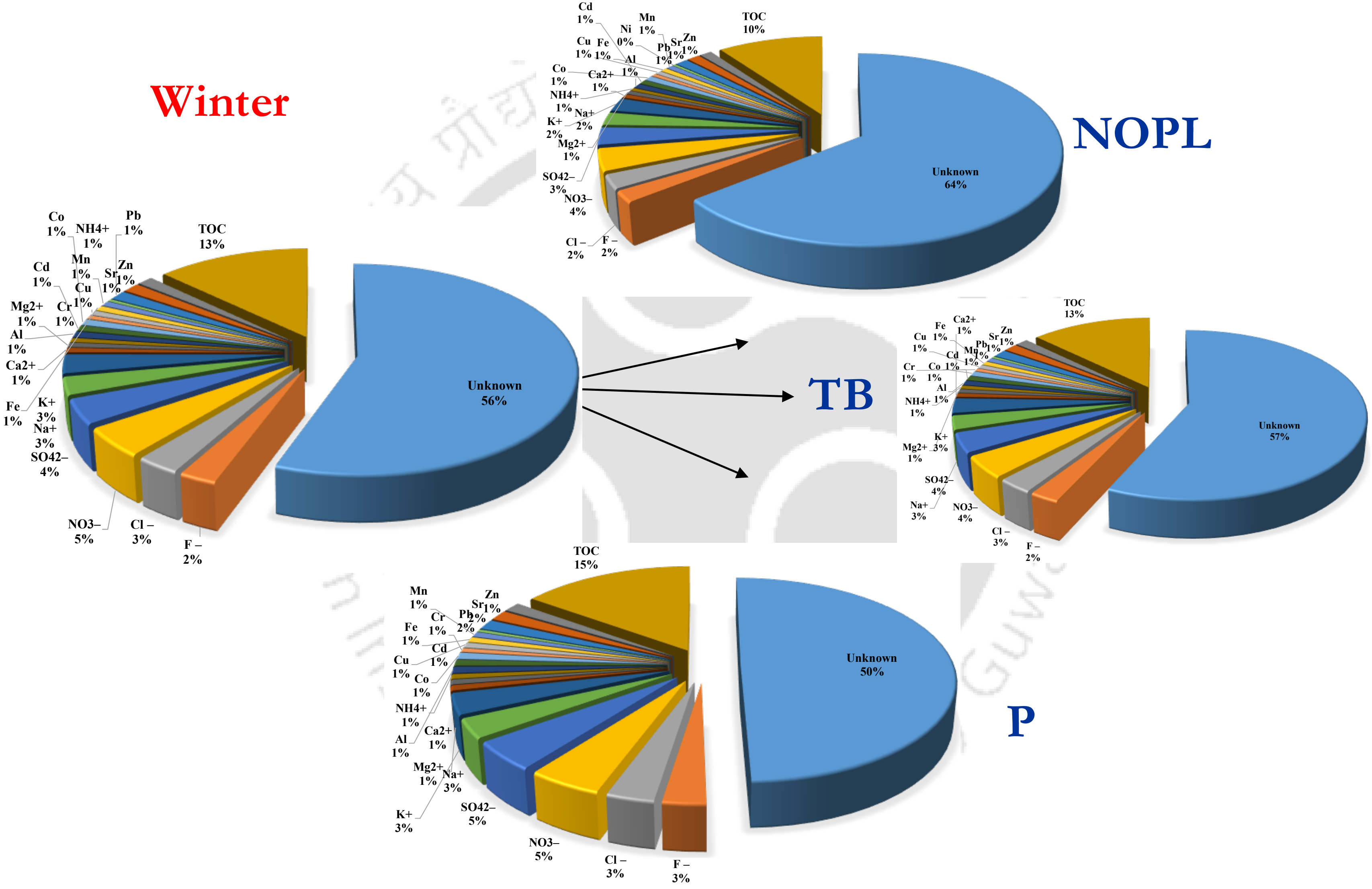

Fig D.10 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S4 during the study period

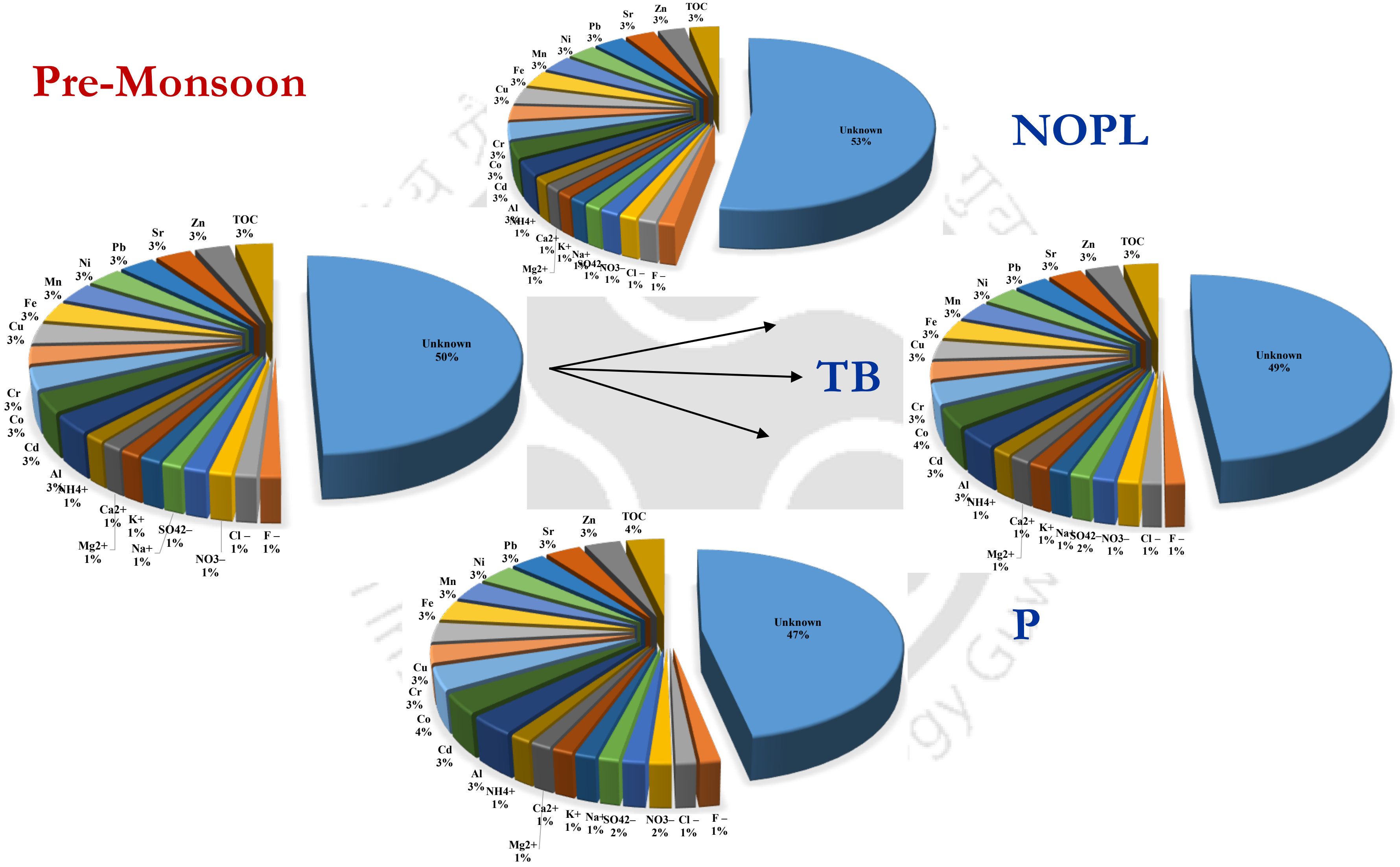

Fig D.11 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S4 during the study period

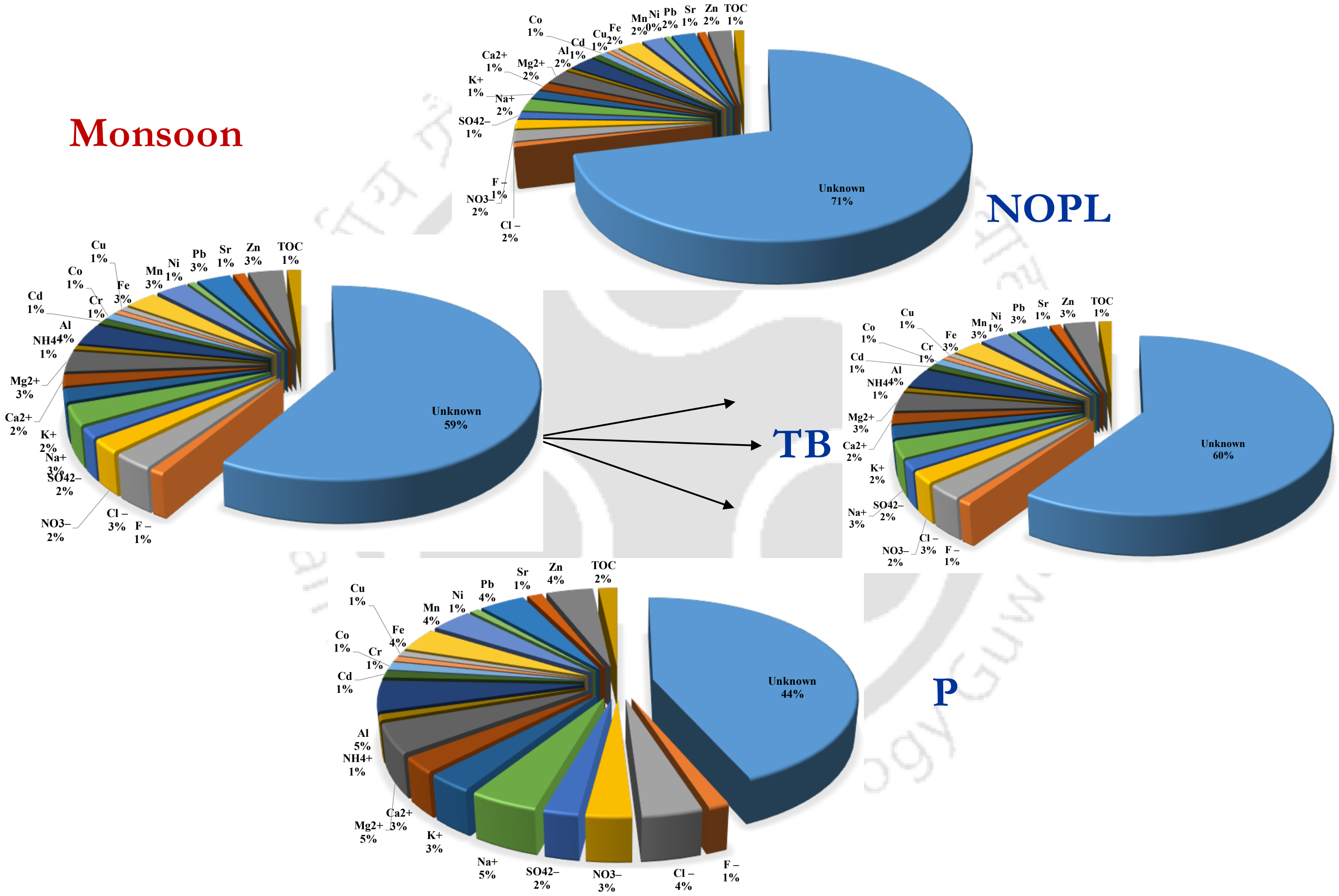

Fig D.12 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S4 during the study period

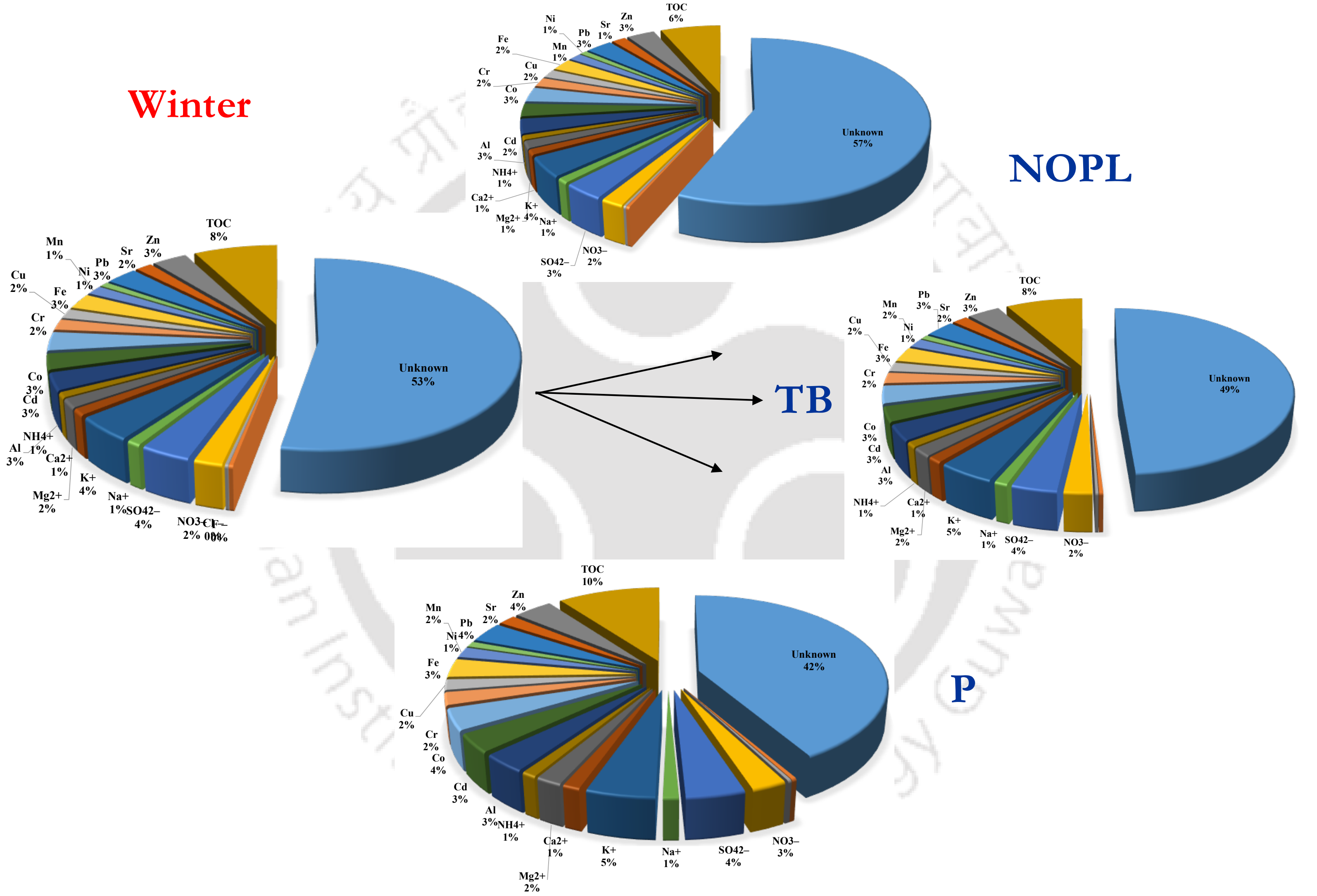

Fig D.13 Chemical components in PM$_{10}$ and in regions of respiratory tract for samples collected in S5 during the study period

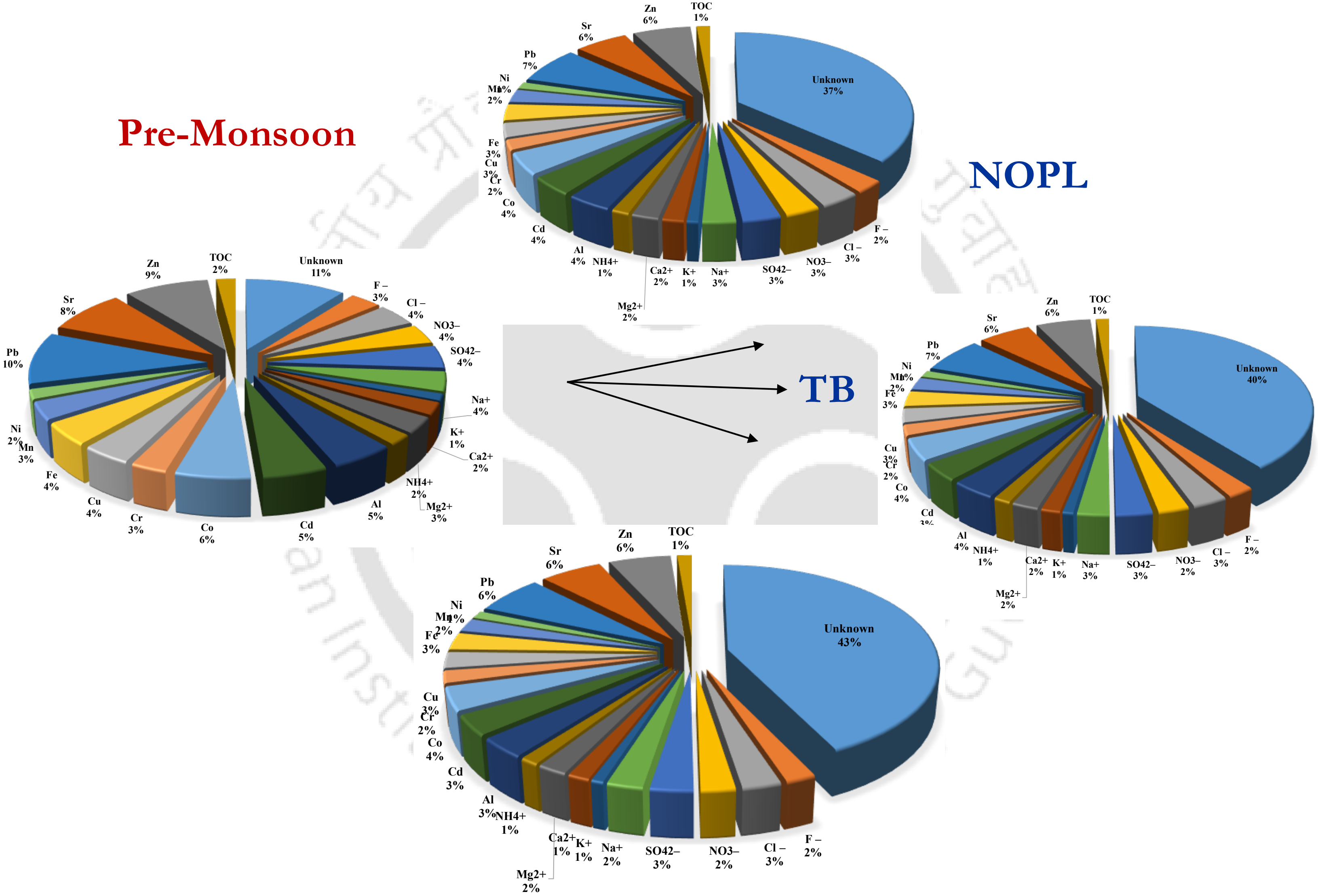

Fig D.14 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S5 during the study period

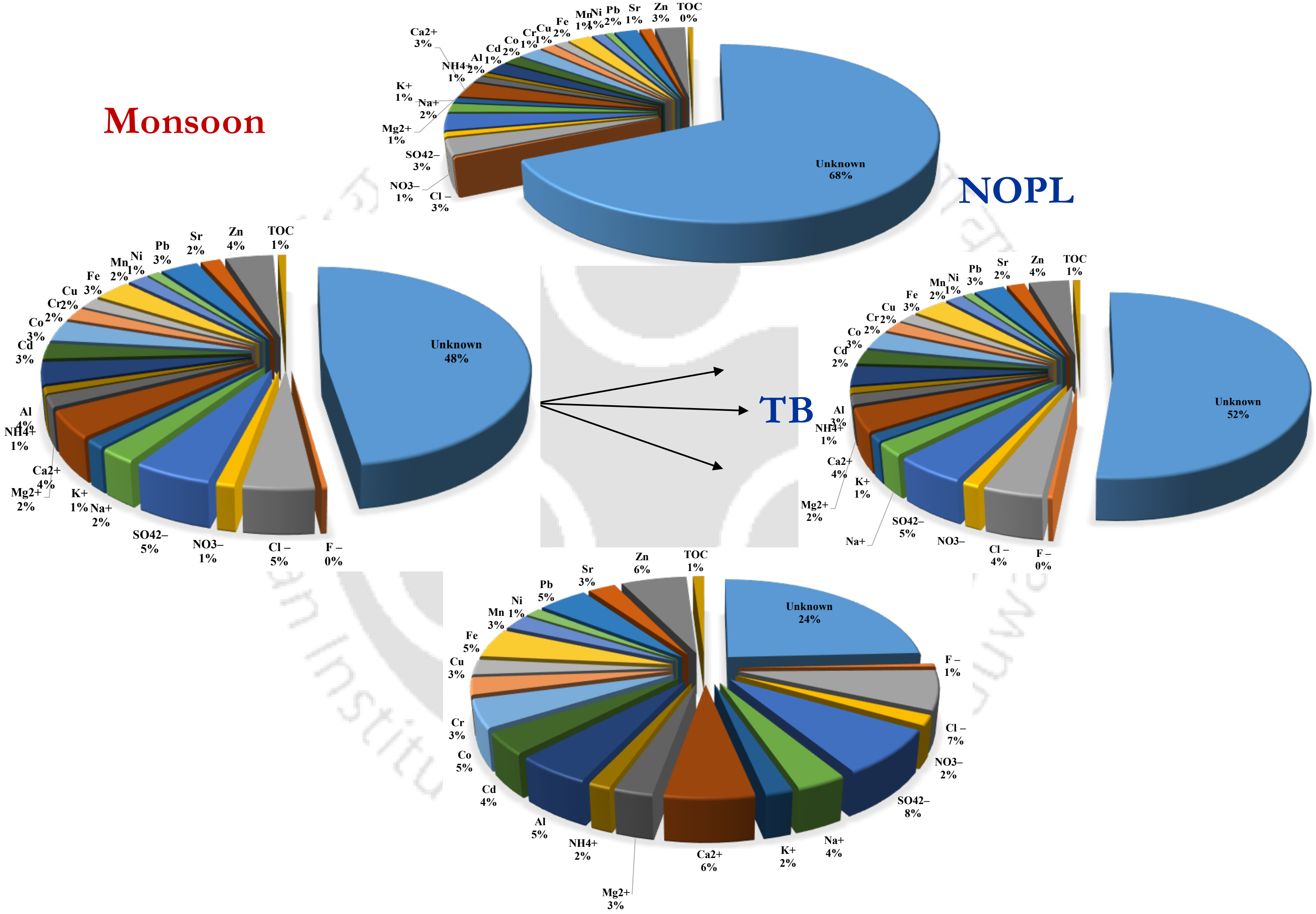

Fig D.15 Chemical components in PM_{10} and in regions of respiratory tract for samples collected in S5 during the study period

PUBLICATIONS

Journal Papers:

1. **Garaga R**, Sahu SK, Kota SH. A Review of Air Quality Modeling Studies in India: Local and Regional Scale. Current Pollution Reports. 2018:1-15.
2. **Garaga R**, Avinash CK, Kota SH. Seasonal variation of airborne allergenic fungal spores in ambient PM10—a study in Guwahati, the largest city of north-east India. Air Quality, Atmosphere & Health. 2018:1-10.
3. Kota SH, Guo H, Myllyvirta L, Hu J, Sahu SK, **Garaga R**, Ying Q, Gao A, Dahiya S, Wang Y, Zhang H. Year-long simulation of gaseous and particulate air pollutants in India. Atmospheric Environment. 2018:244-55.
4. **Garaga R**, Kota SH. Characterization of PM10 and impact on human health during the annual festival of lights (Diwali). Journal of Health and Pollution. 2018 Dec;8(20).
5. Wasim J, Minas I, **Garaga R**, Euripides GS, Kota SH, Ying Q, Guo B. Source apportionment of carbonaceous pollutants in fine and coarse atmospheric particles in Doha, Qatar. Journal of the Air & Waste Management Association, 2019, 69 (11), 1277-1292.
6. **Garaga R,** Chakraborty S, Zhang H, Gokhale S, Qiao X, Kota SH. Influence of anthropogenic emissions on wet deposition of pollutants and rainwater acidity in Guwahati, a UNESCO heritage city in Northeast India. Atmospheric Research. 2020 Feb 1; 232:104683.

7. **Garaga R**, Gokhale S, Kota SH. Source Apportionment of Size-Segregated Atmospheric Particles and the Influence of Particles Deposition in the Human Respiratory Tract in Five Locations in Assam, India. (Draft under preparation)

International Conferences:

1. **Garaga R**, Kota SH. Characterization of PM10 and its impact on human health during annual festival of lights (Diwali) in Northeast India. The American Geophysical Union (AGU) Fall Meeting, Sanfrancisco, USA, 2016.
2. **Garaga R**, Sahu SK, Ramavathu SK, Kota SH. Studying Air Quality During Fireworks in a College Campus. First Indian International Conference on Air Quality Management (IICAQM), Chennai, India, 2016.
3. **Garaga R**, Sahu, SK, Kota SH. Chemical composition and sources to rain water in north-east India. Asia Oceania Geosciences Society (AOGS) 15th Annual Meeting, Hawaii, USA, 2018.
4. **Garaga R**, Gokhale S, Kota SH. Source apportionment of size-segregated atmospheric particles in different locations in largest city of North-East India. The American Geophysical Union (AGU) Fall Meeting, Washington, D.C, USA, 2018.
5. Sahu S, **Garaga R**, Chejarla V, Guo H, Bharti R, Zhang H, Jianlin H, Ying Q, Kota SH. Predicting PM2.5 concentration and associated mortality across India based on GAM model using aerosol optical depth and meteorological parameters. The American Geophysical Union (AGU) Fall Meeting, Washington, D.C, USA, 2018.
6. **Garaga R**, Gokhale S, Kota SH. Source Apportionment of Size-Segregated Atmospheric Particles and Influence of Particles Deposition in Human Respiratory Tract in Five Locations in Assam, India. China-India Association of Indian Scientists (CIAAS), Delhi, India, 2019.

7. **Garaga R**, Gokhale S, Kota SH. Source Apportionment of Size-Segregated Atmospheric Particles and Influence of Particles Deposition in Human Respiratory Tract in Five Locations in Assam, India. Asia Oceania Geosciences Society (AOGS) 16th Annual Meeting, Singapore, 2019.

AWARDS/ACHIEVEMENTS

2019	***Best Paper Award*** in *China-India Association of Indian Scientists* (**CIAAS**), *International Conference* in Delhi, India.
2018	***Young researcher's Travel Grant*** by Department of Science & Technology **(DST)**, India to attend *Centennial AGU Fall meeting 2018, International Conference* in Washington, D.C, USA.
2018	***Student Travel Assistance Fund*** **(STAF*)*** by IIT Guwahati for attending the *AGU Fall meeting 2018 International Conference*
2016	***Student Travel Grant*** by IIT Guwahati for attending the 1st Indian International Conference on Air Quality Management (**IICAQM**), IIT Chennai, India
2015-present	***Ministry of Human Resources Development (MHRD) scholarship*** for Doctoral research at IIT Guwahati