Current atmospheric aerosol research in India

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Atmospheric aerosols are important from a perspective of ambient air pollution and health to humans and other biological receptors as well as for potential effects on local weather and global climate. This review attempts to account for the different research efforts of individual research groups and regulatory agencies in India on the issue of atmospheric aerosols and their effects. The review refers to representative studies reporting the physical characterization (size), chemical composition (organic and inorganic), radiative forcing effects and health effects of aerosols. There are several reports on source apportionment studies identifying sources of aerosols and some focus on specific issues. The review also points out a significant need for more data with a greater spatial and temporal resolution for better understanding of the dynamics of atmospheric aerosols in the Indian context.

Keywords: Aerosols, air pollution, health effects, research efforts.

Introduction

ATMOSPHERIC aerosols are broadly defined as dispersions (or suspensions) of solid or liquid in the atmosphere. The term particulate matter (PM) is commonly used to represent the solid phase suspended matter in the atmosphere. In environmental engineering literature, the term aerosol and PM are often interchangeably used even though the term aerosol has a broader definition and scope. These aerosols are derived from a wide range of natural and anthropogenic sources on earth and within the atmosphere. The study of aerosols in the atmosphere from an environmental and public perspective has been in vogue for the last several decades and has been brought into the focus especially due to effects that are global in scale. The effects of atmospheric aerosols are also closely linked with the fate and transport of gas-phase components in the atmosphere. Some of the fundamental issues underlying the behaviour of atmospheric aerosols and their interaction with gas-phase components and the surrounding environment are compiled in some excellent treatises, which are now widely used as textbook material in courses related to atmospheric aerosols and chemistry[1–4]. In addition, there are a number of reviews on the current trends in aerosol science as applicable to processes in the atmosphere. Some of these reviews focus on specific issues of atmospheric aerosols. These include the characterization of organic components in atmospheric aerosols[5,6], on-line aerosol measurements techniques[7], chemistry of secondary aerosol formation[8], interaction of natural aerosols with the planetary system[9], impact on regional weather[10–14], natural aerosols[15], and surface reactivity and chemistry of aerosols[16].

The study of atmospheric aerosols has two common general objectives: (i) direct impact on public health as a result of exposure near the surface of earth and (ii) role in the atmospheric chemical and physical processes and their consequent possible effects on local and global climate. For either of these, the first step is to gather a large volume of physical data related to the ambient atmosphere. In its simplest form, this data may be the concentration of aerosols, their size and composition as a function of location and time. Time-series datasets can give valuable information about the seasonal variation and also the statistical variability of this data as a function of local meteorology. Direct interpretation of the concentrations can determine if the levels of aerosols in ambient air meet or exceed the ambient regulatory standards. The temporal scale of the time-series data also determines the applicability of the data for specific objectives. Real-time or near-real-time data is useful in gathering information about events related to the fate and transport that occur at very short timescales, whereas time-averaged data can give information that is useful for a different set of data objectives such as receptor exposure levels. In combination with knowledge of pollution sources and characteristics and meteorology in a given region, one can also determine the relative contribution (source apportionment) of various known sources in the region. If this information is statistically defensible, it may be used to formulate or modify public policy and regulation. In general, the pattern of aerosol behaviour or dynamics observed in one part of the world may also be observed in a different part of the world. However, often there are specific regional patterns that may be useful to understand the local behaviour of aerosols (and atmospheric pollutants, in general) and provide valuable insight into the processes affecting local and regional pollutant dynamics.

Following this approach, there have been a number of studies on atmospheric aerosols in India. This review aims at highlighting some of the important aspects that

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have been addressed in studies conducted in India. In this process, the attempt is also to point out some critical gaps in data and analysis with reference to the study of aerosols in the Indian context. The review is arranged on the basis of some of the points raised above and discusses some of the works conducted in the last decade or so.

Ambient aerosol measurements

The measurement of aerosols in the ambient environment can be accomplished with a variety of objectives which define the scope, methodology and analysis of the data collected. The following discussion looks at some of the data collected in different campaigns.

Size-based PM measurements

The primary interest in these measurements is to monitor the mass concentrations of suspended matter in ambient air as a direct indicator of the potential hazard to human health. This is also the most commonly found atmospheric aerosol data in the literature on ambient air. Currently regulatory agencies all over the world have designated several particle sizes and issued ambient air-quality standards. These are PM_{10} and $PM_{2.5}$, where the subscript refers to the particle size represented as an aerodynamic diameter (in microns) cut-off limit. For example, PM_{10} refers to a concentration of PM with an aerodynamic diameter lesser than 10 μm. These are also based on the penetrability of certain particle sizes into the respiratory system. Evidence of this is obtained either through direct physiological observation or inferred from epidemiological data correlating PM exposure to health effects.

Currently the regulatory standards, for PM_{10} and $PM_{2.5}$ in ambient environments in India are 100 and 60 $\mu g/m^3$ respectively[17]. Additionally, another representation of comparison with regulatory standards is the air quality index (AQI), which is a colour-coded tool that is intended to indicate to the general public the extent to which the ambient air quality has exceeded the regulatory limit. The colours green, yellow, orange and red represent good air quality, moderate air quality, unhealthy for sensitive groups and unhealthy for all groups. For $PM_{2.5}$, concentrations of 0–15.4 $\mu g/m^3$ is considered to be good (green); 15.4–35.5 $\mu g/m^3$ is considered to be moderate (yellow); 35.5–55.4 $\mu g/m^3$ is considered to be unhealthy for sensitive groups (orange) and 55.4–140.4 $\mu g/m^3$ is considered as unhealthy for all groups (red). Concentrations exceeding 210 $\mu g/m^3$ are considered to be hazardous. Correspondingly the AQI is also indicated numerically as 0–50 for good, 51–100 for moderate, 101–150 for unhealthy for sensitive groups, 151–200 for unhealthy for all groups and > 300 for hazardous rating.

One of the most commonly used method to collect PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ is the time-averaged method using a high or low volume sampler which intercepts the desired size PM on a filter medium. The mass collected on the filter medium is measured gravimetrically using a 4-digit or 7-digit balance. The reported values of PM require the flow rate to be measured as well. Different impactors and flow rates are used for PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ collection. A variation of the PM_{10} sampler is the respirable dust sampler (RDS) that uses a mini-cyclone instead of an impactor plate (as in the case of PM samplers) to achieve the desired size cut-off, and is designated as a respirable suspended PM. While the gravimetric instruments mentioned above provide physical data measurements directly in terms of mass, the time-averaging that is required to observe the mass collected on the filter media requires relatively high sampling time ranging from 4 to 24 h for ambient air-quality monitoring. Time-averaged measurements are useful in evaluating general trends in ambient air quality in a given region or a season, and for comparing trends between regions. Multichannel speciation samplers are also used to collect PM samples, especially when a multi-component chemical analysis of the sample is the objective. Up to four channels of PM samplers are operated simultaneously with different filter media suitable for the analysis of different chemical species.

Lower timescale measurements are useful in evaluating specific events in an environment. To accomplish this, there are other instruments that use indirect methods to estimate PM concentrations. One of these is the beta-gauge monitor which uses a signal attenuation between a beta radiation source and a detector with the collection of PM on a filter paper. The size cut-off is still achieved using an impactor plate at the inlet of the sampling device. The sensitivity of the measurement requires a lower mass on the filter paper and consequently, the sampling time is much lower compared to gravimetric measurements using traditional PM samplers. A calibration between the attenuated signal and actual mass concentration must be obtained. An oscillating microbalance technique is also used in the measurement of PM.

A very popular technique of measurement of PM is using an optical method of laser scattering. These instruments can be operated in a mode to collect a single particle size or multiple size ranges continuously. The output is a number concentration that is converted to a mass concentration using some information or assumptions about particle shape and density. These instruments are useful for measuring almost real-time analysis of PM. Typically, there is a filter at the exit of the air stream in the instrument to physically collect the PM. This filter can be analysed to check for the integrated mass measurement or for chemical analysis.

A particle size distribution is sometimes desired to evaluate the size characteristics in a specific environ-

ment. This may be ambient air or specific occupational environment. The most commonly used size-fractionating equipment is the cascade impactor. This works on the same principle as the regular PM samplers, but has a series of impactors and collection surfaces which allow the classification of different sizes. The range of sizes depends on the number and type of impactors. Typically the size fractions reported are mass fractions and are gravimetric in nature. In this category there are personal cascade impactors that are highly portable and have miniaturized impactors and collection plates.

In general, PM in the atmosphere can be classified on the basis of their mode of formation as coarse, accumulation and nuclei[3]. Coarse-mode particles are generated from activities that cause resuspension of solids from the soil or other surface materials due to wind or vehicle-induced turbulence and erosion or attrition arising from the contact of two solid surfaces in a process, such as movement of vehicle tyres on road surfaces. Coarse-mode aerosols are also generated from the drying of sea spray consisting on salt particles. In these cases, the PM that is suspended in the atmosphere has the primary composition of the material that was present as a solid (or aqueous droplet in the case of sea spray) in the environment. The nuclei-mode particles are formed by the condensation of material present in the vapour phase to form a solid phase, or on the surface of an exiting solid-phase nucleus. The accumulation-mode particles are formed from the aggregation of nuclei-mode particles and are not large enough to settle easily in the atmosphere, and therefore, have a very long lifetime in the atmosphere compared to the coarse-mode particles. Generally, the transition between the accumulation mode and the coarse mode is between 1 and 3 μm, whereas the nuclei mode starts around 10 nm.

Based on the above definition, PM_{10} is considered as coarse mode, whereas $PM_{2.5}$ is considered as fine particulates and in accumulation mode. Ultrafine PM is generally designated as PM below an aerodynamic diameter of about 300 nm (ref. 3). Currently there are no regulatory guidelines or standards for ultrafine PM in ambient air. Particles in the nanometer size range are formed in vehicular exhaust and grow to larger sizes due to condensation of other gases in the exhaust or in the atmosphere as the exhaust cools away from the tailpipe of the vehicle. At this size range it is more practical to express the PM magnitude as a number concentration rather than as a mass concentration. The most commonly used instrument in this category is the scanning mobility particle sizer (SMPS). This instrument consists of two segments – a particle-classifying device and a size-measuring device. The particle-classifying device is usually a differential mobility analyser (DMA), which uses the electrical mobility of particles to separate them and then a condensation particle counter (CPC) is used to count the particle by an optical method. These instruments also provide almost real-time data with reasonably low particle losses. These instruments have been used in a wide variety of scenarios to understand the fate and transport of ultrafine PM from different sources.

Field measurements: The regulatory agencies that have the primary task of enforcing environmental quality have also taken the responsibility of measuring these parameters on a regular basis. The Central Pollution Control Board (CPCB) has embarked upon a very large programme, National Air Quality Monitoring Programme (NAMP), to monitor and report some of the priority air pollutants to the general public. Under this programme, there are a large number of monitoring stations all across the country. Details of this programme are available at CPCB website[17]. Some of these stations have been continuously monitoring the pollutants and are equipped with instrumentation that can provide very short timescale data. The PM monitors in these automated stations are the beta-gauge attenuation monitors. The data from these monitoring stations are reported at the NAMP website. In addition to CPCB, the State Pollution Control Boards also monitor the air quality in different cities and display their data at several locations, or in local newspapers in a city for the benefit of the general public. The PM data that are currently collected include the total suspended particulate matter (TSPM), which has a cut-off of around 100 μm, PM_{10}, $PM_{2.5}$ and respirable suspended particulate matter (RSPM). The CPCB has also recently completed an exhaustive six-city source apportionment study. The reports pertaining to this study are available at the CPCB website[18]. The study was conducted in six cities – Bengaluru, Chennai, Delhi, Kanpur, Mumbai and Pune. As part of this study, a large amount of data was collected at seven different locations in a each city corresponding to kerbside, industrial and residential sites, and in three different seasons for a period of at least 20 days at each location and season. PM_{10} and $PM_{2.5}$ were collected along with RSPM and TSPM. Though the focus of the study was source apportionment, the large dataset that exists can also be used potentially for other analyses as well.

One of the major source of ambient aerosols in an urban environment is vehicles of different types[19]. The primary emissions from vehicles result from the combustion of different types of fuel – petrol, diesel, liquid petroleum gas (LPG) or compressed natural gas (CNG). Changes in fuel types are usually based on a large ambient aerosol measurement campaign conducted locally or adapted from observations elsewhere in a different environment. An example of this is the change in the policy to convert most public transport vehicles in New Delhi to CNG from petrol or diesel based on studies of the ambient aerosol data[20,21]. Measurements that were conducted after a significant amount of time after the policy implementation indicate the changes in aerosol concentrations

brought about by the policy change[22–25]. Though these studies do not unequivocally determine the effectiveness of the policy change, the data from these reports provide useful insights into the complexity of the problem. For instance, one of the offsetting factors for change in fuel type is the increase in the number of vehicles on the road since the policy change. Exposure to aerosols from other household combustion activities such cooking with biofuels or wood in rural or semi-urban areas is a matter of concern from a personal health perspective.

There have been a number of studies that look at the exposure to aerosols and related constituents for people engaged in cooking with solid biofuels. In a study conducted in approximately 400 rural homes in Andhra Pradesh, the concentration of PM_{10} was found to be significantly higher for wood and dung compared to gas and kerosene. The mean 24 h PM_{10} concentrations were in the range 73–732 $\mu g/m^3$, with the lower end of the range attributed to gas fuels[26]. In similar studies reported in Tamil Nadu, the average PM_{10} concentrations were reported as 500–2000 $\mu g/m^3$ (ref. 27). In this study, it was also reported that the estimated 24 h exposure level from biofuel burning was in the range 231 ± 109 $\mu g/m^3$ for those involved in cooking and 90 ± 21 $\mu g/m^3$ for those not involved in cooking, but in the same household. In a study to measure fine PM indoors in 11 households at two urban centres in India, particle number concentrations of more than 300,000 cm^{-3} and mass concentrations of more than 1000 $\mu g\ m^{-3}$ were detected. These were attributed to the biofuels and the poor ventilation of kitchens in most of these cases[28]. In another study, it was found that the particle size of aerosols associated during the cooking process was in the accumulation mode in the 0.1–0.3 μm size range, in contrast to indoor PM in the 1.0–2.0 μm size range during the non-cooking period. During the frying process, the droplet was found to be coarser in the 0.7–1.0 μm range (ref. 29). In this context, there have been a few collaborative initiatives with several prominent industrial houses and individuals who have come up with ideas of novel cooking stoves that reduce the amount of exposure by improving the combustion process. Studies of this nature are also useful in directing public policy in terms of investment in the development of better combustible biofuels or in the design of efficient and inexpensive stoves. Field sampling of cookstove emissions was conducted in two rural locations in India, where $PM_{2.5}$, particulate surface area concentration in both tracheobronchial and alveolar regions along with carbon monoxide (CO) were measured in 120 households and two roadside restaurants[30]. Novel indices for the performance of traditional and improved stoves were evaluated and presented in this study. Other studies correlate the use of biofuels and stoves to decreased lung function[31,32].

There are a number of studies that have observed time series of ambient PM and ultrafine data at different locations in India. Some of these studies report specific relationships between vehicular traffic patterns and ambient concentrations. In an estimation conducted using published data around 2000, Chennai city emitted about 7 tonnes/day of PM (ref. 33). In most cases, there was a correlation of the ambient measurement at a particular location with traffic flow, composition and intensity parameters[34–40]. Common analysis includes the estimation of the ambient concentrations using emission factors and count data for vehicles. In other interesting studies, the focus has been on the exposure for drivers, especially two-wheelers (scooters and motorcycles) and auto-rickshaws. Measured exposure concentrations for an average of data collected from 60 measurements sets were 190 $\mu g/m^3$ for $PM_{2.5}$, 42 $\mu g/m^3$ for black carbon (BC) and 280×10^3 particles/cm^3 (ref. 41). The in-vehicle concentrations were significantly higher than the ambient measured concentrations for these indices. Mean RSPM levels were measured as 370–2860 $\mu g/m^3$ (ref. 42). Both these studies were conducted in different locations in Delhi. This information is very useful for the design of protection devices for two-wheelers and also other transportation design aspects, such as designating lanes for different vehicles. This information on two-wheelers is also applicable to cars in India, especially the small-sized ones. The transport of aerosol and gas-phase pollutants in the microenvironments of auto-rickshaws and cars with open windows would be useful information.

There are a vast number of studies that look at different types of applications, which are difficult to categorize. Nevertheless, these are valuable data that may be useful in gaining better understanding of the processes. A few studies look at the contributions of ultrafine PM in ambient aerosols. In addition to presenting the mass and number concentrations, this also presents the correlation between both, since mass concentration measurements are relatively cheaper. The study reports that the correlation fails after the mass concentration increases beyond 300 $\mu g/m^3$ possibly, beyond which it decreases[20,43]. Aerosol concentrations were measured in a ostensibly pristine environment such as the Himalayas. In the Kullu–Manali region, measurement of 24 h average value of ultrafine particles of three size ranges is 18045 ± 1212, 16811 ± 2790 and 15407 ± 3109 N/cm^3 respectively, indicating an influence of traffic[44]. In a much more remote Himalayan site, the 1 h-average total number concentration varied between 220 and 27300 N/cm^3, indicating transport of fine particulates from other locations[45]. There are other measurements that show size distribution and composition[46–50] in various urban scenerios, in urban commercial activities such as shipping harbours[51], and agricultural activities[52]. Another focus of aerosol and air-quality measurement studies is to investigate the impact on historical monuments that are important tourist centres, as is the case with the Taj Mahal in Agra[53].

Among the India-centric events that generate significant amount of aerosol in the urban environment is the festival of Diwali. There are a number of studies that have measured emission from a variety of firecrackers during an intense period of combustion activities at different levels on and off the ground. A study conducted in Lucknow in 2005 showed 24 h average PM_{10} concentration for Diwali day to be 753 $\mu g/m^3$, which was 2.5–5.7 times higher than the average pre-Diwali measurement. Also the 12 h average night-time value of PM_{10} was 1206 $\mu g/m^3$, which was four times higher than the daytime 12 h average[54]. In another study in Delhi, the 24 h average PM_{10} measurement was 317.2–616.8 $\mu g/m^3$ (ref. 55). Another study measured the concentration of metals in the aerosols as a result of fireworks in Hyderabad[56] and Delhi[57]. There are other local festivals that involve burning of fireworks, but on a smaller scale than is the case with Diwali.

The apparent increase in the number of incidences of dense fog formation (especially in North India) causes severe disruption of commercial activity, air traffic and in some severe cases, even road and rail traffic. Fog forms on an aerosol seed that then allows condensation of water vapour for the growth of the particle until the size and the water content become sufficiently large to cause a reduction in visibility. If an increase in the intensity and incidence of fog is observed, it is possible that the increase is triggered by the increased presence of hygroscopic aerosols in the atmosphere. Since this is of great interest in some parts of the country, several studies have focused on this aspect[58–61]. These studies have looked at the correlation between the aerosols observed, and the formation, intensity and characteristics of the fog water (chemical constituents).

Chemical composition of aerosols

The understanding of the chemical composition of atmospheric aerosols is important for three main reasons: (i) better understanding about the possible sources of the aerosol in the atmosphere, (ii) basis for the hypothesis of various atmospheric chemical and physical processes and (iii) information regarding the potential toxicity and health impacts from specific constituents of aerosols. The chemical composition therefore plays a critical role in the determination of the final size of the aerosol during its residence time in the atmosphere.

The type of instrumentation that is used for the analysis of chemical composition depends on the type of species that one is interested in probing. PM samplers used for the collection of aerosols for mass concentration can be used for a limited set of analysis. Standard methods exist for the analysis of each of the organic and inorganic constituents associated with aerosols. The United States Environmental Protection Agency (USEPA) has a good compendium of recommended analytical methods for each class of chemical[62] and in the EPA-SW 846 series of methods.

Inorganic elements: Metals associated with aerosols, for most part, are formed from attrition and therefore of the coarse mode. There are cases of very high temperature processes where the metal is molten or vapourized and then cooled to form condensed aerosol particles. There are a number of studies that have measured metal concentrations in an urban ambient atmosphere for a various end-uses[63–72]. The six-city source apportionment study conducted by the CPCB has large datasets of metals associated with aerosols[18]. These studies look at the average metal concentration in aerosols from activities that are sustained and therefore represent a near-steady-state input to the lower atmosphere, where most of the human exposure occurs. These sources are usually a combination of vehicles, commercial and construction/demolition activities. These measurements also showcase the applicability of a variety of analytical instruments available for analysis of including atomic absorption spectroscopy (AAS), inductively coupled plasma–atomic emission spectroscopy (ICP-AES), inductively coupled plasma mass spectrometry (ICPMS), X-ray fluorescence and scanning electron microscopy (SEM) using energy or wavelength dispersive spectra (EDS or WDS). As mentioned earlier, during Diwali, the aerosol concentrations are high and large fractions of these aerosols are metal-based particles that impart the different colours[54–57]. There are other studies that look at specific environments and the levels of aerosol concentration in them such as indoor–outdoor exchange[73], mercury emissions from power plants[74], and measurements in pristine environments indicating long-range transport of pollutants from nearby polluting regions[75].

Ions: A large number of studies focus on the presence of ions in the aerosols. These ions can also indicate the source of a particular chemical constituent and therefore the process that contributes to the source. Some ions are absorbed or adsorbed in the aerosol in their original form, whereas some undergo transformation on the surface of the aerosol, especially in the presence of water. Sea-water spray contains ions in solution and produce salt aerosols as the water evaporates. In relatively humid regions, water condensation can occur on certain types of aerosols, thus contributing to the ionic chemistry associated with the aerosol[1]. Campaigns looking at an overall ionic composition in urban areas are spread all over the country as in the case of the metals[67,71,76–85]. These analyses are usually done with the extraction of the ions from the filter media (Nylon or Teflon) with water followed by analysis of the cations and anions separately using ionchromatography. The six-city source apportionment study conducted by the CPCB has large datasets of ions associated

with aerosols[18]. There are reports of alternative analytical methods such as the one using Raman spectroscopy[85]. Ionic chemistry occurs at very short timescales compared to the time-averaged filter-based collection of aerosols. Since all these measurements are not on-line or real-time, they provide information regarding the average state in most cases. Some studies look at the ionic composition in pristine environments such as the Himalayas, again either highlighting a background natural concentration of ions or long-range transport of aerosols[86,87]. There are some reports studying interaction of ions leading to the transformation or growth of aerosols and their potential impact on the environment[88–90]. There are a number of studies that focus specifically on the chemistry of a particular ion or a group of ions that pertain to a certain process. Ions of sulphur and nitrogen are some that have been prominent in studies[53,91–94]. These studies have great significance in the chemical cycling in local environments. An example of ionic chemistry in aerosols is in fog droplets. A large amount of literature is available studying the chemistry of fog water, where interactions are possible between the seed aerosol–water and the water–air interfaces. A few studies in India have focused on the ion chemistry of fog waters as well[60,61].

Polyaromatic hydrocarbons: These constitute a class of compounds that take prominence in aerosol organic composition as they are formed from combustion of many common solid fossil fuels and wood. Some of these are potential carcinogens and owing to their hydrophobicity, they tend to be present on PM suspended in the atmosphere. A number of studies have looked at the monitoring of polyaromatic hydrocarbons (PAHs) in the atmosphere in an urban environment[95–97], interaction between gas phase and PM[98], emissions during specific events[54], and atmospheric deposition of PAHs[99]. The preferred method for PAH analysis is by extraction of the aerosol collection medium (preferably quartz or glass fibre filter with low organic binder) with an organic solvent such as dichloromethane or hexane followed by sample clean-up and chemical analysis by GC-MS.

Other organic constituents: The chemical analysis with GC–MS of aerosol samples reveals a wide range of chemicals from aliphatic hydrocarbons and derivatives, aromatic compounds and their derivatives, organic acids and esters, alcohols, ketones, aldehydes, amides and other specific compounds. This type of scan of chemicals in PM gives valuable insight to the sources of various chemicals. Organic compounds associated with aerosols are derived from a range of natural and anthropogenic sources. A large number of alkanes and organic acids appear in aerosol samples from natural sources as well. For this reason, the analysis of molecular markers for specific sources is measured to look for signatures of specific sources. A number of studies are exploratory in nature, trying to understand the organic aerosol composition[100–102]. The six-city source apportionment study conducted by the CPCB has large datasets of molecular markers and source profiles associated with aerosols[18]. There are specific markers whose chemical analysis is difficult because they are in trace levels and therefore require more expensive methods of analysis. One such chemical species are the dioxins, which are formed from combustion of specific solid waste components[103]. One of the significant components of this type of molecular marker analysis is the chemical characterization of different types of PM identified from different sources. This provides a source profile that will be useful in linking constituents in ambient air to a particular source. Studies to characterize source profiles are also carried out[104]. There are other studies that pick out a specific species from a group of species identified in an attempt to look for dependencies on other parameters in the environment[101,102,105–108].

Black carbon or elemental carbon: This species of elemental or black carbon (EC/BC) is one of the highly measured components in the atmosphere. There are two applications of the EC/BC data: (i) in estimating the relative contribution of sources that generate BC (primarily soot production from combustion sources) and (ii) their impact on radiative forcing and the consequent impact on regional weather patterns. One of the instrumental techniques available for the analysis of carbon on aerosols is the use of thermal/optical reflectance carbon analyser developed by the Desert Research Institute (DRI), Reno, USA. One of the protocols developed by DRI[109,110], analyses the organic carbon (OC) fraction and EC using a combination of temperature programming and optical reflectance measurements of PM on a quartz filter-paper sample. The OC/EC ratio is often used to crudely characterize if the aerosol sample collected is dominated by a combustion source, such as vehicles. A large set of OC/EC/BC data is available for a number of locations in India, pertaining to both urban and rural environments[111–120]. In addition, the six-city source apportionment study conducted by the CPCB has large datasets of OC/EC measurements associated with urban aerosols[18]. Most of these measurements are conducted using the DRI-OC/EC analyser.

Source apportionment

Closely linked to the chemical composition and total mass measurements of aerosols in the environment is the problem of apportioning source of these aerosols to obtain relevant public policy for the management of ambient air quality. The apportionment of chemical constituents to a source requires the knowledge of chemical constituent targets, possible sources of these targets, source profiles

(or chemical characterization of the source with respect to each chemical constituent of interest), emission inventory and ambient concentration of these target chemical constituents. Standard regulatory models such as the chemical mass balance (CMB) model are available to estimate the relative contribution of each of the known sources. This sort of analysis also gives some insight into sources that might not have been taken into account. In addition, other data-processing models are available and have been developed that obtain correlations between observed data and possible sources. These include positive matrix factorization (PMF), principal component analysis (PCA) and other statistical receptor models. These models estimate the distribution of the components among one class of aerosols whose mass concentration measurement is available. For example, the source attribution of PM_{10} class of particulates. A number of source apportionment studies have been reported in the Indian context. Some of these look at the general urban scenario[121–127], others look at the methodologies of the source estimation and the modification[128–131]. The six-city source apportionment study conducted by the CPCB applies different techniques of source apportionment to the large sets of data collected in association with urban aerosols[18]. Source apportionment is a difficult task since the open atmospheric environment is complex and the analysis is fraught with uncertainties. Chemical analysis of specific signatures can be useful in getting specific information about sources, if it is possible to obtain reliable data.

Effect on climate

One of the main focal points in recent decades has been the effect of air pollution on regional weather patterns and global effects such as greenhouse gas effect. One of the key components in this discussion is the presence of aerosols, especially BC on radiative forcing or altering the amount of radiation received at a particular location on the earth's surface. There are a number of uncertainties in this regard, and consequently, there are global inter-institutional programmes to measure the effects of radiative forcing by aerosols[132,133]. Regional interest is primarily from a point of view of prediction of the weather patterns, especially the monsoons on which India depends critically. Reports of local weather changes due to large aerosol clouds lead to questions of pollution sources, local regulation and management[57]. A large number of independent and cooperative studies have looked at aerosols in general and specific constituents in the atmosphere[134–142]. One of the instruments used for this purpose is the aethalometer, which is an optical device that measures the intensity of aerosols. Some of the measurements relate to an aerosol optical depth (AOD) as an indicator of the aerosol. Energy budget over the earth's surface is calculated using these measurements and estimations of their composition. Some specific studies look at changes in optical properties in the atmosphere as a result of specific events, such as large-scale combustion burning[143].

Health effects

Health effects arising from exposure to PM or aerosols have been widely reported and vary from mild respiratory ailments to very severe chronic effects. The health effects attributed to aerosols are mostly assessed by epidemiological studies from a small set of population or from hospital data of symptoms linked to occupational information of the patients. There are a few examples of studies where direct physiological evidence (whether in humans or laboratory animals) is available for connecting aerosol exposure to an ailment. Experiments were done by exposing rats with different doses of PM for different intervals of time. Post-mortem analysis of the rats revealed an increase in relative lung weight and inflammatory changes[144]. The respiratory health of workers employed in a municipal solid-waste disposal landfill facility was monitored as a function of age, gender and socio-economic conditions. In comparison to a control group, the landfill workers had a higher incidence of symptoms such as respiratory trouble and a host of other ailments. Spirometry tests revealed impairment in lung function for a significant section of these people. Sputum cytology studies revealed other pathological evidences of deposition, inflammation or infection on different components of the respiratory system[145]. Epidemiological studies have also been conducted to correlate exposure to health effects. These usually require a large amount of data and a statistical model for the correlation. One study looks at a time-series analysis of short-term exposure of PM_{10} on mortality rates. It was found that there was an increase of 0.44% in mortality rate per 10 pg/m^3 increase in daily average PM_{10} concentration[146].

Spirometry tests for lung function were conducted on school children ranging in age between 9 and 17 in Delhi. Lung function was reduced by 43.5% in children in the test group compared to 25.7% in the control group. Using statistical models, PM_{10} in children was found to be associated with restrictive, obstructive or combined-type lung function deficits[147]. Bioaerosol samples in the vicinity of wastewater treatment plants indicated higher biological particles in the air, including endotoxins and bacteria. Workers in this wastewater treatment plant displayed symptoms of respiratory disorders, gastrointestinal tract infections, fatigue and headache. These symptoms are typical of the response to endotoxins. The role of endotoxins associated with wastewater treatment plant emissions in inflammatory response was studied[148]. A study was also conducted to investigate the effect of indoor air pollution in buildings in New Delhi. Respiratory health

effects and sick-building syndrome on occupants living in the inefficiently designed buildings with poor ventilation were studied. The statistical study concluded that women and children are more at risk compared to men. High sick-building syndrome scores were reported in these households due to poor design and the use of biofuels indoors[149].

Other studies simulating the effect of PM on tissue or surface in the respiratory tract were also performed. One such study simulated the effect of the particles emitted from biofuel combustion the activity of surfactants present on the surface of the lungs and in other parts of the respiratory tract. Experimental studies were performed using model surfactants similar to those present in the lungs and it was found that particles from wood combustion increased the minimum surface tension on the surfactant, indicating a dysfunction and a greater tendency of alveolar collapse *in vivo* leading to respiratory distress[150]. In another study, the efficiency of particle deposition in the respiratory tract as the function of soluble component present in the particle was simulated using a numerical model. This study indicated that particles can also grow up to twice their size due to water uptake during their travel in the respiratory channels[151].

There have been a number of studies that measure the exposure of different groups of population to PM as a function of occupation, location and socio-economic background[26,27,29–31]. In combination with the health-effects studies, these exposure studies are also valuable in designing better tools for combustion, or better residential design, or a change in public policy.

Conclusions

A large body of aerosol work in India is focused on the radiative forcing effect and on the effect on climate in the region. There is also a reasonably large and growing set of studies involved in aerosol characterization. Thought this is done in a sporadic manner at different locations, it has covered wide sections of the country. There has been some research on specific issues that do not come under the purview of ambient air quality, but those which focus on the processes and mechanisms underlying the cause of pollution. Though the general principles are known through previous studies conducted elsewhere and available in the open literature, these focused studies help in validating these principles in the specific cases as well as identifying issues that are very specific to India.

In comparison to the United States, the amount of ambient environmental data associated with air pollution is relatively sparse. There have been a few large-scale field campaigns for specific goals, with some success. The biggest need at this time is the collection of reliable data with good spatial and temporal resolution. The regulatory bodies such as CPCB and state PCBs have set out in this direction and have embarked on a large-scale automated gathering of data, which are expected to be in the public domain and available for the integration of other supporting studies. A large number of mechanistic information is available from field and laboratory studies in the open literature. This information is used for most part by regulatory agencies and researchers in India. However, there are specific cases which are India-centric and cannot be obtained by extrapolation of observations elsewhere. Therefore, more abundant field data are required. Data can be used to model and validate scenarios of pollutant behaviour in response to changes in policy or pollution generation patterns.

Key to obtaining large datasets is the increase and upgradation of instrumentation for on-line analysis of aerosols and pollutants in general. A big difference between the state of the science in aerosol literature in general and the studies in India is the current reliance on techniques of chemical characterization that are not real-time. One of the problems this poses is the lack of resolution of chemical trends that are short-term in scale. Therefore, it is possible to miss the trend of chemical reactions in the environment. There are a few ultrafine PM monitors that have been used and reported in this review. On-line chemical composition is a significant challenge. One such example of a sophisticated on-line analysis tool is the aerosol mass spectrometer (AMS). The AMS is an instrument that was developed to study real-time analysis of organic species in aerosols along with particle size distributions[152]. There are several reviews of the application of this instrument in the literature in different parts of the world. In the detailed literature review that was conducted, the use of AMS was not found in India. A related research and academic gap is the development of analytical equipment specific to aerosol research. A significant indicator is the number of courses on aerosol physics and chemistry offered across the country. Aerosol science as an environmental science application academic discipline is yet to be airborne in India. There needs to be a greater thrust on academic courses focusing on aerosol chemistry and physics in the environmental engineering curriculum.

General pollutant transport theories are well established and validated all over the world. There are, however, very specific local microenvironments such as that in a busy urban road, where pollutant transport assessment can lead to better prediction of exposure and risk. Field experimental validation of such studies is challenging and can be substituted by conducting studies in the simulated environments such as wind tunnels. The simulation and experimental validation of a large number of such studies can be useful in the formulation of efficient strategies of optimized, environmentally safe modes of transportation.

1. Seinfeld, J. H. and Pandis, S. N., *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, Wiley Interscience, Hoboken, NJ, USA, 2006, 2nd edn.
2. Finlayson-Pitts, B. and Pitts Jr, J., *Atmospheric Chemistry: Fundamentals and Experimental Techniques*, John Wiley, USA, 1986.

3. Hinds, W. C., *Aerosol Technology: Properties, Behavior and Measurement of Airborne Particles*, Wiley-Interscience, USA, 1999, 2nd edn.
4. Friedlander, S. K., *Smoke, Dust and Haze: Fundamentals of Aerosol Dynamics*, Oxford University Press, USA, 2000, 2nd edn.
5. Jacobson, M. C., Hansson, H.-C., Noone, K. J. and Charlson, R. J., Organic atmospheric aerosols: review and state of the science. *Rev. Geophys.*, 2000, **38**(2), 267–294.
6. Jimenez, J. L. *et al.*, Evolution of organic aerosols in the atmosphere. *Science*, 2009, **326**(5959), 1525–1529.
7. Sullivan, R. C. and Prathers, K. A., Recent advances in our understanding of atmospheric chemistry and climate made possible by online aerosol analysis instrumentation. *Anal. Chem.*, 2005, **77**, 3861–3886.
8. Kroll, J. H. and Seinfeld, J. H., Chemistry of secondary organic aerosol: formation and evolution of low-volatility organics in the atmosphere. *Atmos. Environ.*, 2008, **42**(16), 3593–3264.
9. Carslaw, K. S., Boucher, O., Spracklen, D. V., Mann, G. W., Rae, J. G. L., Woodward, S. and Kulmala, M., A review of natural aerosol interactions and feedbacks within the Earth system. *Atmos. Chem. Phys.*, 2010, **10**(4), 1701–1737.
10. Menon, S., Koch, D., Beig, G., Sahu, S., Fasullo, J. and Orlikowski, D., Black carbon aerosols and the third polar ice cap. *Atmos. Chem. Phys.*, 2010, **10**(10), 4559–4571.
11. Menon, S., Hansen, J., Nazarenko, L. and Luo, Y., Climate effects of black carbon aerosols in China and India. *Science*, 2002, **297**(5590), 2250–2253.
12. Reddy, M. S. and Venkataraman, C., Atmospheric optical and radiative effects of anthropogenic aerosol constituents from India. *Atmos. Environ.*, 2000, **34**(26), 4511–4523.
13. Adams, P. J., Seinfeld, J. H. and Koch, D. M., Global concentrations of tropospheric sulfate, nitrate, and ammonium aerosol simulated in a general circulation model. *J. Geophys. Res. D*, 1999, **104**(11), 13791–13823.
14. Massie, S. T., Torres, O. and Smith, S. J., Total ozone mapping spectrometer (TOMS) observations of increases in Asian aerosol in winter from 1979 to 2000. *J. Geophys. Res. D*, 2004, **109**(18), D18211/1–D18211/14.
15. Carslaw, K. S., Boucher, O., Spracklen, D. V., Mann, G. W., Rae, J. G. L., Woodward, S. and Kulmala, M., A review of natural aerosol. *Atmos. Chem. Phys*., 2010, **10**(4), 1701–1737.
16. Al-Abadleh, H. A. and Grassian, V., Oxide surfaces as environmental interfaces. *Surf. Sci. Rep.*, 2003, **52**, 63–161.
17. Central Pollution Control Board (CPCB), India, National Air Monitoring Programme (NAMP), New Delhi; http://www.cpcb.nic.in/air.php
18. CPCB, Source apportionment studies; http://www.cpcb.nic.in/Source_Apportionment_Studies.php
19. Ramachandra, T. V. and Shwetmala, Emission from India's transport sector: statewise synthesis. *Atmos. Environ.*, 2009, **43**(34), 5510–5517.
20. Monkkonen, P. *et al.*, Relationship and variations of aerosol number and PM10 mass concentrations in a highly polluted urban environment, New Delhi, India. *Atmos. Environ.*, 2004, **38**(3), 425–433.
21. Laakso, L. *et al.*, Aerosol particles in the developing world, a comparison between New Delhi in India and Beijing in China. *Air, Water Soil Pollut.*, 2006, **173**(1–4), 5–20.
22. Chelani, A. B. and Devotta, S., Impact of change in fuel quality on PM10 in Delhi. *Bull. Environ. Contam. Toxicol.*, 2005, **75**(3), 600–607.
23. Reynolds, C. C. O., Grieshop, A. P. and Kandlikar, M., Climate and health relevant emissions from in-use Indian three-wheelers fueled by natural gas and gasoline. *Environ. Sci. Technol.*, 2011, **45**(6), 2406–2412.
24. Gupta, I., Salunkhe, A. and Kumar, R., Modelling 10-year trends of PM10 and related toxic heavy metal concentrations in four cities in India. *J. Hazard. Mater.*, 2010, **179**(1–3), 1084–1095.
25. Khillare, P. S., Agarwal, T. and Shridhar, V., Impact of CNG implementation on PAHs concentration in the ambient air of Delhi: a comparative assessment of pre- and post-CNG scenario. *Environ. Monitor. Assess.*, 2003, **147**(1–3), 223–233.
26. Balakrishnan, K., Sambandam, S., Ramaswamy, P., Mehta, S. and Smith, K. R., Exposure assessment for respirable particulates associated with household fuel use in rural districts of Andhra Pradesh, India. *J. Exposure Anal. Environ. Epidemiol.*, 2004, **14**(Suppl. 1), S14.
27. Balakrishnan, K. *et al.*, Daily average exposures to respirable particulate matter from combustion of biomass fuels in rural households of southern India. *Environ. Health Perspect.*, 2002, **110**(11), 1069–1075.
28. Moenkkoenen, P. *et al.*, Fine particle number and mass concentration measurements in urban Indian households. *Sci. Total Environ.*, 2005, **347**(1–3), 131–147.
29. Varghese, S., Gangamma, S., Patil, R. and Sethi, V., Particulate respiratory dose to Indian women from domestic cooking. *Aerosol. Sci. Technol.*, 2005, **39**(12), 1201–1207.
30. Sahu, M., Peipert, J., Singhal, V., Yadama, G. N. and Biswas, P., Evaluation of mass and surface area concentration of particle emissions and development of emissions indices for cookstoves in rural India. *Environ. Sci. Technol.*, 2011, **45**(6), 2428–2434.
31. Padhi, B. K. and Padhy, P. K., Domestic fuels, indoor air pollution, and children's health: the case of rural India. *Ann. N.Y. Acad. Sci.*, 2008, **1140**, 209–217.
32. Smith, K. R., Aggarwal, A. L. and Dave, R. M., Air pollution and rural biomass fuels in developing countries: a pilot village study in India and implications for research and policy. *Atmos. Environ.*, 1983, **17**(11), 2343–2362.
33. Nesamani, K. S., Estimation of automobile emissions and control strategies in India. *Sci. Total Environ.*, 2010, **408**(8), 1800–1811.
34. Majumdar, D., Mukherjee, A. K. and Sen, S., Apportionment of sources to determine vehicular emission factors of BTEX in Kolkata, India. *Water, Air Soil Pollut.*, 2009, **201**(1–4), 379–388.
35. Barman, S. C., Singh, R., Negi, M. P. S. and Bhargava, S. K., Fine particles (PM2.5) in residential areas of Lucknow city and factors influencing the concentration. *Clean: Soil, Air, Water*, 2008, **36**(1), 111–117.
36. Ahmad, Y., Bhargava, R. and Parida, M., Transport generated air pollution on national highways in Haridwar district. *J. Inst. Public Health Eng., India*, 2005, **4**, 15–18.
37. Mohanraj, R., Azeez, P. A. and Pattabhi, S., Automobile pollution in urban Coimbatore, India. *Nature, Environ. Pollut. Technol.*, 2005, **4**(4), 621–626.
38. Gokhale, S. B. and Patil, R. S., Size distribution of aerosols (PM10) and lead (Pb) near traffic intersections in Mumbai (India). *Environ. Monitor. Assess.*, 2004, **95**(1–3), 311–324.
39. Raj, P. E. *et al.*, Influence of urban activity on columnar aerosols, ozone and precipitable water in the city of Pune, India. *Pollut. Res.*, 2002, **21**(3), 381–391.
40. Srimuruganandam, B. and Shiva Nagendra, S. M., Characteristics of particulate matter and heterogeneous traffic in the urban area of India. *Atmos. Environ.*, 2011, **45**(18), 3091–3102.
41. Apte, J. S. *et al.*, Concentrations of fine, ultrafine, and black carbon particles in auto-rickshaws in New Delhi, India. *Atmos. Environ.*, 2011, **45**(26), 4470–4480.
42. Saksena, S., Prasad, R. K. and Shankar, V. R., Daily exposure to air pollutants in indoor, outdoor and in-vehicle microenvironments: a pilot study in Delhi. *Indoor Built Environ.*, 2007, **16**(1), 39–46.
43. Moenkkoenen, P., Koponen, I. K., Lehtinen, K. E. J., Haemeri, K., Uma, R. and Kulmala, M., Measurements in a highly polluted Asian mega city: observations of aerosol number size distribution, modal parameters and nucleation events. *Atmos. Chem. Phys.*, 2005, **5**(1), 57–66.
44. Sharma, N. L., Kuniyal, J. C., Singh, M., Negi, A. K., Singh, K. and Sharma, P., Number concentration characteristics of ultrafine

aerosols (atmospheric nanoparticles/aitken nuclei) during 2008 over western Himalayan region, Kullu-Manali, India. *Indian J. Radio Space Phys.*, 2009, **38**(6), 326–337.
45. Komppula, M., Lihavainen, H., Hyvarinen, A.-P., Kerminen, V.-M., Panwar, T. S., Sharma, V. P. and Viisanen, Y., Physical properties of aerosol particles at a Himalayan background site in India. *J. Geophys. Res. D*, 2009, **114**(12), D12202/1–D12202/11.
46. Venkataraman, C., Reddy, C. K., Josson, S. and Reddy, M. S., Aerosol size and chemical characteristics at Mumbai, India, during the INDOEXIFP (1999). *Atmos. Environ.*, 2002, **36**(12), 1979–1991.
47. Raghavendra Kumar, K. *et al.*, Size segregated mass concentration and size distribution of near surface aerosols over a tropical Indian semi-arid station, Anantapur: impact of long range transport. *Sci. Total Environ.*, 2009, **407**(21), 5589–5604.
48. Shandilya, K. K., Khare, M. and Gupta, A. B., Suspended particulate matter distribution in rural–industrial Satna and in urban–industrial South Delhi. *Environ. Monit. Assess.*, 2007, **128**(1–3), 431–445.
49. Gupta, S., Srivastava, A. and Jain, V. K., Particle size distribution of aerosols and associated heavy metals in kitchen environments. *Environ. Monit. Assess.*, 2008, **142**(1–3), 141–148.
50. Sharma, M. and Maloo, S., Assessment of ambient air PM10 and PM2.5 and characterization of PM10 in the city of Kanpur, India. *Atmos. Environ.*, 2005, **39**(33), 6015–6026.
51. Joseph, J., Patil, R. S. and Gupta, S. K., Estimation of air pollutant emission loads from construction and operational activities of a port and harbour in Mumbai, India. *Environ. Monit. Assess.*, 2009, **159**(1–4), 85–98.
52. Mittal, S. K., Singh, N., Agarwal, R., Awasthi, A. and Gupta, P. K., Ambient air quality during wheat and rice crop stubble burning episodes in Patiala. *Atmos. Environ.*, 2009, **43**(2), 238–244.
53. Kulshrestha, U. C., Kumar, N., Saxena, A., Kumari, K. M. and Srivastava, S. S., Identification of the nature and source of atmospheric aerosols near the Taj Mahal (India). *Environ. Monit. Assess.*, 1995, **34**(1), 1–11.
54. Barman, S. C., Singh, R., Negi, M. P. S. and Bhargava, S. K., Ambient air quality of Lucknow City (India) during use of fireworks on Diwali Festival. *Environ. Monit. Assess.*, 2008, **137**(1–3), 495–504.
55. Sarkar, S., Khillare, P. S., Jyethi, D. S., Hasan, A. and Parween, M., Chemical speciation of respirable suspended particulate matter during a major firework festival in India. *J. Hazard. Mater.*, 2010, **184**(1–3), 321–330.
56. Kulshrestha, U. C., Nageswara Rao, T., Azhaguvel, S. and Kulshrestha, M. J., Emissions and accumulation of metals in the atmosphere due to crackers and sparkles during Diwali festival in India. *Atmos. Environ.*, 2004, **38**(27), 4421–4425.
57. Singh, D. P. *et al.*, Study of temporal variation in ambient air quality during Diwali festival in India. *Environ. Monit. Assess.*, 2010, **169**(1–4), 1–13.
58. Mehta, B., Venkataraman, C., Bhushan, M. and Tripathi, S. N., Identification of sources affecting fog formation using receptor modeling approaches and inventory estimates of sectoral emissions. *Atmos. Environ.*, 2009, **43**(6), 1288–1295.
59. Mohan, M. and Payra, S., Influence of aerosol spectrum and air pollutants on fog formation in urban environment of megacity Delhi, India. *Environ. Monit. Assess.*, 2009, **151**(1–4), 265–277.
60. Safai, P. D. *et al.*, Aerosol characteristics during winter fog at Agra, North India. *J. Atmos. Chem.*, 2008, **61**(2), 101–118.
61. Lakhani, A., Parmar, R. S., Satsangi, G. S. and Prakash, S., Chemistry of fogs at Agra, India: influence of soil particulates and atmospheric gases. *Environ. Monit. Assess.*, 2007, **133**(1–3), 435–445.
62. United States Environmental Protection Agency (USEPA) – Technology Transfer Network: Ambient Monitoring Technology Information Center (TTN/AMTIC); http://www.epa.gov/ttn/amtic/methods.html, last updated January 2011.
63. Kar, S., Maity, J. P., Samal, A. C. and Santra, S. C., Metallic components of traffic-induced urban aerosol, their spatial variation, and source apportionment. *Environ. Monit. Assess.*, 2010, **168**(1–4), 561–574.
64. Kulshrestha, A., Satsangi, P. G., Masih, J. and Taneja, A., Metal concentration of PM2.5 and PM10 particles and seasonal variations in urban and rural environment of Agra, India. *Sci. Total Environ.*, 2009, **407**(24), 6196–6204.
65. Chelani, A. B., Gajghate, D. G. and Hasan, M. Z., Atmospheric toxic metal concentrations in urban area of Nagpur city. *Indian J. Environ. Protect.*, 2001, **21**(3), 250–257.
66. Vijayanand, C., Rajaguru, P., Kalaiselvi, K., Selvam, K. P. and Palanivel, M., Assessment of heavy metal contents in the ambient air of the Coimbatore city, Tamil Nadu, India. *J. Hazard. Mater.*, 2008, **160**(2–3), 548–553.
67. Samanta, G. *et al.*, Air pollution in Calcutta during winter – a three year study. *Curr. Sci.*, 1998, **75**(2), 123–138.
68. Singh, R., Barman, S. C., Negi, M. P. S. and Bhargava, S. K., Metals concentration associated with respirable particulate matter (PM10) in industrial area of Eastern UP India. *J. Environ. Biol.*, 2008, **29**(1), 63–68.
69. Basha, S. *et al.*, Assessment of heavy metal content in suspended particulate matter of coastal industrial town, Mithapur, Gujarat, India. *Atmos. Res.*, 2010, **97**(1–2), 257–265.
70. Vijay Bhaskar, B., Jeba Rajasekhar, R. V., Muthusubramanian, P. and Kesarkar, A. P., Ionic and heavy metal composition of respirable particulate in Madurai, India. *Environ. Monit. Assess.*, 2010, **164**(1–4), 323–336.
71. Tiwari, K., Pandey, A. and Pandey, J., Atmospheric deposition of heavy metals in a seasonally dry tropical urban environment (India). *J. Environ. Res. Dev.*, 2008, **2**(4), 605–611.
72. Yadav, S. and Rajamani, V., Air quality and trace metal chemistry of different size fractions of aerosols in N–NW India – implications for source diversity. *Atmos. Environ.*, 2006, **40**(4), 698–712.
73. Srivastava, A. and Jain, V. K., Relationships between indoor and outdoor air quality in Delhi. *Indoor Built Environ.*, 2003, **12**(3), 159–165.
74. Salve, P. R., Krupadam, R. J. and Wate, S. R., Assessment of atmospheric particulate mercury Hg(p) in the vicinity of coal based power plant. *J. Indian Assoc. Environ. Manage.*, 2006, **33**(1), 39–41.
75. Cong, Z., Kang, S., Dong, S., Liu, X. and Qin, D., Elemental and individual particle analysis of atmospheric aerosols from high Himalayas. *Environ. Monit. Assess.*, 2010, **160**(1–4), 323–335.
76. Kulshrestha, A., Bisht, D. S., Masih, J., Massey, D., Tiwari, S. and Taneja, A., Chemical characterization of water–soluble aerosols in different residential environments of semi arid region of India. *J. Atmos. Chem.*, 2009, **62**(2), 121–138.
77. Verma, S. K., Deb, M. K., Suzuki, Y. and Tsai, Y. I., Ion chemistry and source identification of coarse and fine aerosols in an urban area of eastern central India. *Atmos. Res.*, 2010, **95**(1), 65–76.
78. Rao, P. M., Rao, K. G., Rama Rao, D. V., Reddy, M. K. and Rao, M. V., Studies on quantification of air borne particulates and acidic ions in the environment around a coal mining area. *Indian J. Environ. Prot.*, 1998, **18**(6), 443–450.
79. George, S. K., Nair, P. R., Parameswaran, K., Jacob, S. and Abraham, A., Seasonal trends in chemical composition of aerosols at a tropical coastal site of India. *J. Geophys. Res. D*, 2008, **113**(16), D16209/1–D16209/15.
80. Madhavan, B. L., Niranjan, K., Sreekanth, V., Sarin, M. M. and Sudheer, A. K., Aerosol characterization during the summer monsoon period over a tropical coastal Indian station, Visakhapatnam. *J. Geophys. Res. D*, 2008, **113**(21), D21208/1–D21208/16.
81. Behera, S. N. and Sharma, M., Investigating the potential role of ammonia in ion chemistry of fine particulate matter formation for

an urban environment. *Sci. Total Environ*., 2010, **408**(17), 3569–3575.
82. Ragini, N., Chandrashekara, M. S., Nagaiah, N. and Paramesh, L., Study of atmospheric electrical conductivity, SO_2, NO_2, aerosols SPM (> 10 μ) and RSPM (< 10 μ) in Mysore city, India. *Toxicol. Environ. Chem.*, 2009, **91**(4), 605–609.
83. Singh, R., Sharma, B. S. and Chalka, S. N., Seasonal air quality profile of inorganic ionic composition of PM10 near Taj Mahal in Agra, India. *Environ. Monit. Assess*., 2010, **168**(1–4), 195–203.
84. Harpale, V. M., Ralegankar, S. D., Jadhav, D. B. and Dhole, S. D., Chemical identification of urban air particulates in the environment of Pune City (India) by Raman spectroscopy. *Aerosol Air Qual. Res*., 2006, **6**(3), 295–304.
85. Rastogi, N. and Sarin, M. M., Long-term characterization of ionic species in aerosols from urban and high-altitude sites in western India: role of mineral dust and anthropogenic sources. *Atmos. Environ*., 2005, **39**(30), 5541–5554.
86. Ahmad, S. and Hasnain, S. I., Chemical characteristics of atmospheric precipitation at high Himalaya, Ganga headwater, India. *J. Appl. Geochem*., 2006, **8**(2), 148–153.
87. Chatterjee, A. *et al.*, Aerosol chemistry over a high altitude station at northeastern Himalayas, India. *PLoS One*, 2010, **5**(6), DOI:10.1371/journal.pone.0011122.
88. Joshi, S. D., Pandya, G. H., Phadke, K. M., Tajne, D. S., Jain, A. K., Gajrani, C. P. and Yennawar, P. K., An investigation into the acid content of aerosols in the ambient air at the Taj Mahal, Agra. *Environ. Pollut.*, 1989, **58**(2–3), 87–96.
89. Kulshrestha, U. C., Reddy, L. A. K., Satyanarayana, J. and Kulshrestha, M. J., Real-time wet scavenging of major chemical constituents of aerosols and role of rain intensity in Indian region. *Atmos. Environ*., 2009, **43**(32), 5123–5127.
90. Chate, D. M. and Murugavel, P., Atmospheric aerosol formation and its growth during the cold season in India. *J. Earth Syst. Sci*., 2010, **119**(4), 471–477.
91. Safai, P. D., Budhavant, K. B., Rao, P. S. P., Ali, K. and Sinha, A., Source characterization for aerosol constituents and changing roles of calcium and ammonium aerosols in the neutralization of aerosol acidity at a semi-urban site in SW India. *Atmos. Res*., 2010, **98**(1), 78–88.
92. Pandey, S. K., Tripathi, B. D., Mishra, V. K. and Prajapati, S. K., Size fractionated speciation of nitrate and sulfate aerosols in a sub-tropical industrial environment. *Chemosphere*, 2006, **63**(1), 49–57.
93. Tripathi, B. D., Chaturvedi, S. S. and Tripathi, R. D., Seasonal variation in ambient air concentration of nitrate and sulfate aerosols in a tropical city, Varanasi. *Atmos. Environ*., 1996, **30**(15), 2773–2778.
94. Venkataraman, C., Sinha, P. and Bammi, S., Sulphate aerosol size distributions at Mumbai [Bombay], India, during the INDOEX-FFP (1998). *Atmos. Environ*., 2001, **35**(15), 2647–2655.
95. Santosh, H. S., Nandini, N., Pandit, G. G. and Sharma, S., Quantification of polycyclic aromatic hydrocarbons in size fractionated ambient aerosol and their source identification using wind profile data. *Asian J. Microbiol., Biotechnol. Environ. Sci.*, 2007, **9**(4), 855–860.
96. Taneja, A., Saini, R. and Masih, A., Indoor air quality of houses located in the urban environment of Agra, India. *Annu. NY Acad. Sci.*, 2008, **1140**, 228–245.
97. Ravindra, K., Wauters, E., Tyagi, S. K., Mor, S. and Grieken, R., Assessment of air quality after the implementation of compressed natural gas (CNG) as fuel in public transport in Delhi, India. *Environ. Monit. Assess*., 2006, **115**(1–3), 405–417.
98. Venkataraman, C., Thomas, S. and Kulkarni, P., Size distributions of polycyclic aromatic hydrocarbons–gas/particle partitioning to urban aerosols. *J. Aerosol. Sci*., 1999, **30**(6), 759–770.
99. Sahu, S. K., Pandit, G. G. and Puranik, V. D., Dry deposition of polycyclic aromatic hydrocarbons associated with atmospheric particulate matters in an urban site, Mumbai, India. *Aerosol Air Qual. Res.*, 2008, **8**(4), 437–446.
100. Fu, P. Q., Kawamura, K., Pavuluri, C. M., Swaminathan, T. and Chen, J., Molecular characterization of urban organic aerosol in tropical India: contributions of primary emissions and secondary photooxidation. *Atmos. Chem. Phys*., 2010, **10**(6), 2663–2689.
101. Gupta, A. K., Nag, S. and Mukhopadhyay, U. K., Characterisation of PM10, PM2.5 and benzene soluble organic fraction of particulate matter in an urban area of Kolkata, India. *Environ. Monit. Assess*., 2006, **115**(1–3), 205–222.
102. Manuel, J. A., Phadke, K. M. and Kumar, A., Soluble organic fraction and benzo-apyrene in particulate matter at kerbside and ambient air. *Chem. Environ. Res.*, 2004, **13**(3&4), 227–231.
103. Lal, R. B. *et al.*, Levels of polychlorinated dibenzodioxins and dibenzofurans (PCDDs/Fs) in airborne particulate matter collected in Delhi, India. *Org. Compd.*, 2007, **69**, 700/1–700/4.
104. Sharma, M., Agarwal, A. K. and Bharathi, K. V. L., Characterization of exhaust particulates from diesel engine. *Atmos. Environ*., 2005, **39**(17), 3023–3028.
105. Rengarajan, R., Sudheer, A. K. and Sarin, M. M., Aerosol acidity and secondary organic aerosol formation during wintertime over urban environment in western India. *Atmos. Environ*., 2011, **45**(11), 1940–1945.
106. Kawamura, K. and Pavuluri, C. M., New directions: need for better understanding of plastic waste burning as inferred from high abundance of terephthalic acid in South Asian aerosols. *Atmos. Environ*., 2010, **44**(39), 5320–5321.
107. Pavuluri, C. M., Kawamura, K. and Swaminathan, T., Water-soluble organic carbon, dicarboxylic acids, ketoacids, and α-dicarbonyls in the tropical Indian aerosols. *J. Geophys. Res. D*, 2010, **115**(D11), D11302/1–D11302/15.
108. Miyazaki, Y., Aggarwal, S. G., Singh, K., Gupta, P. K. and Kawamura, K., Dicarboxylic acids and water-soluble organic carbon in aerosols in New Delhi, India, in winter: characteristics and formation processes. *J. Geophys. Res. D*, 2009, **114**(19), D19206/1–D19206/12.
109. Chow, J. C., Watson, J. G., Pritchett, L. C., Pierson, W. R., Frazier, C. A. and Purcell, R. G., The drithermal/optical reflectance carbon analysis system: description, evaluation and applications in US air quality studies. *Atmos. Environ*., 1993, **27**(8), 1185–1201.
110. Chow, J. C. *et al.*, Refining temperature measures in thermal/optical carbon analysis. *Atmos. Chem. Phys. Discuss*., 2005, **5**(4), 4477–4505.
111. Beegum, S. N. *et al.*, Spatial distribution of aerosol black carbon over India during pre-monsoon season. *Atmos. Environ.*, 2009, **43**(5), 1071–1078.
112. Safai, P. D., Kewat, S., Praveen, P. S., Rao, P. S. P., Momin, G. A., Ali, K. and Devara, P. C. S., Seasonal variation of black carbon aerosols over a tropical urban city of Pune, India. *Atmos. Environ*., 2007, **41**(13), 2699–2709.
113. Ram, K. and Sarin, M. M., Spatio-temporal variability in atmospheric abundances of EC, OC and WSOC over northern India. *J. Aerosol Sci*., 2010, **41**(1), 88–98.
114. Ram, K., Sarin, M. M. and Tripathi, S. N., A 1 year record of carbonaceous aerosols from an urban site in the Indo-Gangetic Plain: characterization, sources, and temporal variability. *J. Geophys. Res. D*, 2010, **115**(24), D24313/1–D24313/14.
115. Kumar, K. R. *et al.*, Characterization of aerosol black carbon over a tropical semi-arid region of Anantapur, India. *Atmos. Res*., 2011, **100**(1), 12–27.
116. Rengarajan, R., Sarin, M. M. and Sudheer, A. K., Carbonaceous and inorganic species in atmospheric aerosols during wintertime over urban and high-altitude sites in North India. *J. Geophys. Res. D*, 2007, **112**(21), D21307/1–D21307/16.
117. Chowdhury, Z. *et al.*, Speciation of ambient fine organic carbon particles and source apportionment of PM2.5 in Indian cities. *J. Geophys. Res. D*, 2007, **112**(15), D15303/1–D15303/14.

118. Parashar, D. C., Gadi, R., Mandal, T. K. and Mitra, A. P., Carbonaceous aerosol emissions from India. *Atmos. Environ.*, 2005, **39**(40), 7861–7871.
119. Baxla, S. P., Roy, A. A., Gupta, T., Tripathi, S. N. and Bandyopadhyaya, R., Analysis of diurnal and seasonal variation of submicron outdoor aerosol mass and size distribution in a northern Indian city and its correlation to black carbon. *Aerosol Air Qual. Res*., 2009, **9**(4), 458–469.
120. Tiwari, S. *et al.*, Black carbon and chemical characteristics of PM10 and PM2.5 at an urban site of North India. *J. Atmos. Chem.*, 2010, **62**(3), 193–209.
121. Sunder Raman, R. and Ramachandran, S., Annual and seasonal variability of ambient aerosols over an urban region in western India. *Atmos. Environ*., 2010, **44**(9), 1200–1208.
122. Sathe, Y., Ayare, A. and Srinikethan, G., Application of US EPA PMF model to source apportionment of trace elements in atmospheric aerosols at Kolhapur, Maharashtra (India). *J. Environ. Res. Dev.*, 2011, **5**(3), 597–607.
123. Chakraborty, A. and Gupta, T., Chemical characterization and source apportionment of submicron (PM1) aerosol in Kanpur region, India. *Aerosol Air Qual. Res.*, 2010, **10**(5), 433–445.
124. Gadkari, N. M. and Pervez, S., Source investigation of personal particulates in relation to identify major routes of exposure among urban residentials. *Atmos. Environ*., 2007, **41**(36), 7951–7963.
125. Srivastava, A. and Jain, V. K., Size distribution and source identification of total suspended particulate matter and associated heavy metals in the urban atmosphere of Delhi. *Chemosphere*, 2007, **68**(3), 579–589.
126. Khare, P. and Baruah, B. P., Elemental characterization and source identification of PM2.5 using multivariate analysis at the suburban site of North-East India. *Atmos. Res*., 2010, **98**(1), 148–162.
127. Kar, S., Maity, J. P., Samal, A. C. and Santra, S. C., Metallic components of traffic-induced urban aerosol, their spatial variation, and source apportionment. *Environ. Monit. Assess*., 2010, **168**(1–4), 561–574.
128. Sunder Raman, R., Ramachandran, S. and Rastogi, N., Source identification of ambient aerosols over an urban region in western India. *J. Environ. Monit.*, 2010, **12**(6), 1330–1340.
129. Bhanuprasad, S. G., Venkataraman, C. and Bhushan, M., Positive matrix factorization and trajectory modelling for source identification: a new look at Indian ocean experiment ship observations. *Atmos. Environ*., 2008, **42**(20), 4836–4852.
130. Verma, S., Venkataraman, C., Boucher, O. and Ramachandran, S., Source evaluation of aerosols measured during the Indian ocean experiment using combined chemical transport and back trajectory modeling. *J. Geophys. Res. D*, 2007, **112**(11), D11210/1–D11210/14.
131. Intergovernmental Panel on Climate Change, Radiative forcing of climate change. In *Climate Change 2001*, Cambridge University Press, New York, 2001.
132. Bates, T. S. *et al.*, Aerosol direct radiative effects over the northwest Atlantic, northwest Pacific, and North Indian Oceans: estimates based on *in situ* chemical and optical measurements and chemical transport modelling. *Atmos. Chem. Phys*., 2006, **6**, 1657–1732.
133. Gustafsson, O. *et al.*, Brown clouds over South Asia: biomass or fossil fuel combustion? *Science*, 2009, **323**(5913), 495–498.
134. Raju, M. P., Safai, P. D., Rao, P. S. P., Devara, P. C. S. and Budhavant, K. B., Seasonal characteristics of black carbon aerosols over a high altitude station in Southwest India. *Atmos. Res.*, 2011, **100**(1), 103–110.
135. Babu, S. S., Moorthy, K. K. and Satheesh, S. K., Latitudinal gradient in aerosol properties over the Indian and Southern Oceans during the austral summer. *Curr. Sci*., 2010, **99**(10), 1384–1389.
136. Dey, S. and Tripathi, S. N., Aerosol direct radiative effects over Kanpur in the Indo-Gangetic basin, northern India: long-term (2001–2005) observations and implications to regional climate. *J. Geophys. Res. D*, 2008, **113**(4), D04212/1–D04212/20.
137. Moorthy, K. K., Satheesh, S. K., Babu, S. S. and Dutt, C. B. S., Integrated campaign for aerosols, gases and radiation budget (ICARB): an overview. *J. Earth Syst. Sci*., 2008, **117**(S1), 243–262.
138. Pandithurai, G., Dipu, S., Dani, K. K., Tiwari, S., Bisht, D. S., Devara, P. C. S. and Pinker, R. T., Aerosol radiative forcing during dust events over New Delhi, India. *J. Geophys. Res. D*, 2008, **113**(13), D13209/1–D13209/13.
139. Ram, K., Sarin, M. M. and Hegde, P., Long-term record of aerosol optical properties and chemical composition from a high-altitude site (Manora Peak) in Central Himalaya. *Atmos. Chem. Phys*., 2010, **10**(23), 11791–11803.
140. Gogoi, M. M., Pathak, B., Moorthy, K. K., Bhuyan, P. K., Babu, S. S., Bhuyan, K. and Kalita, G., Multi-year investigations of near surface and columnar aerosols over Dibrugarh, northeastern location of India: heterogeneity in source impacts. *Atmos. Environ*., 2011, **45**(9), 1714–1724.
141. Kharol, S. K., Badarinath, K. V. S., Sharma, A. R., Kaskaoutis, D. G. and Kambezidis, H. D., Multiyear analysis of Terra/Aqua MODIS aerosol optical depth and ground observations over tropical urban region of Hyderabad, India. *Atmos. Environ*., 2011, **45**(8), 1532–1542.
142. Rana, S., Kant, Y. and Dadhwal, V. K., Diurnal and seasonal variation of spectral properties of aerosols over Dehradun, India. *Aerosol Air Qual. Res*., 2009, **9**(1), 32–49.
143. Vadrevu, K. P., Ellicott, E., Badarinath, K. V. S. and Vermote, E., MODIS derived fire characteristics and aerosol optical depth variations during the agricultural residue burning season, north India. *Environ. Pollut.*, 2011, **159**(6), 1560–1569.
144. Pradhan, A., Waseem, M., Dogra, S., Khanna, A. K. and Kaw, J. L., Alterations in bronchoalveolar lavage constituents, oxidant/antioxidant status, and lung histology following intratracheal instillation of respirable suspended particulate matter. *J. Environ. Pathol., Toxicol. Oncol*., 2005, **24**(1), 19–32.
145. Ray, M. R., Roychoudhury, S., Mukherjee, G., Roy, S. and Lahiri, T., Respiratory and general health impairments of workers employed in a municipal solid waste disposal at an open landfill site in Delhi. *Int. J. Hyg. Environ. Health*, 2005, **208**(4), 255–262.
146. Balakrishnan, K., Ganguli, B., Ghosh, S., Sankar, S., Thanasekaraan, V., Rayudu, V. N. and Caussy, H., Short-term effects of air pollution on mortality: results from a time-series analysis in Chennai, India. Research Report, Health Effects Institute, 2011, vol. 157, pp. 7–44.
147. Siddique, S., Banerjee, M., Ray, M. R. and Lahiri, T., Air pollution and its impact on lung function of children in Delhi, the capital city of India. *Water, Air, Soil Pollut.*, 2010, **212**(1–4), 89–100.
148. Gangamma, S., Patil, R. S. and Mukherji, S., Characterization and proinflammatory response of airborne biological particles from wastewater treatment plants. *Environ. Sci. Technol.*, 2011, **45**(8), 3282–3287.
149. Kulshreshtha, P. and Khare, M., A comparative study of indoor air pollution and its respiratory impacts in Delhi, India. *WIT Trans. Ecol. Environ*., 2010, **136**, 287–296.
150. Kanishtha, T., Banerjee, R. and Venkataraman, C., Effect of particle emissions from biofuel combustion on surface activity of model and therapeutic pulmonary surfactants. *Environ. Toxicol. Pharmacol*., 2006, **22**(3), 325–333.
151. Varghese, S. K. and Gangamma, S., Particle deposition in human respiratory tract: effect of water-soluble fraction. *Aerosol Air Qual. Res*., 2006, **6**(4), 360–379.
152. Jimenez, J. L. *et al.*, Ambient aerosol sampling using the Aerodyne Aerosol Mass Spectrometer. *J. Geophys. Res. D*, 2003, **108**(7), 8425–8437.