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REVIEW

Assessment of atmospheric particulate matter and heavy metals: a critical review

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Abstract This paper gives detailed comprehensive review of atmospheric assessment of particulate matter and heavy metals. Previous research works executed on this subject matter in the past four decades were adequately scrutinized. Various equipments for assessing atmospheric particulate matter and heavy metals were presented. Mathematical modeling equations for source apportionment and characterization, deposition rate prediction and health risk characterization of PM_{10} were also presented. However, the following conclusions were made: (1) there is need for improvement on the mathematical models by reducing the number of assumptions made in developing them. (2) Comparative analysis of concentrations of heavy metals in the atmosphere under the same environment for different methodologies should be executed for accuracy purposes. (3) Cost implication of assessing, monitoring and controlling these unfriendly substances should be examined, and hence, involvement of cost engineers may be of immense help. (4) Further research works should be done on Air-Q 2.2.3 model currently identified as a new methodology for provision of quantitative data on the impact of particulate matter exposure on the health of people. (5) Compliance monitoring networks should be designed to ease data collection for the observables, locations and time periods that allowed receptor models to be applied. (6) There is need for much more research works that enable optimal control and regulation of emission of heavy metals into the atmosphere in order to reduce health effects of these inhalable substances.

Keywords Particulate matter · Heavy metal · Atmospheric assessment · Concentration · Air pollution · Atmospheric deposition

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Introduction

Atmosphere is a gaseous layer surrounding the earth. It is the transparent layer through which life-sustaining solar radiation passes and reaches the earth's surface or into the water (Critchfield 1987). However, the emergence of particulate matter and heavy metals due to industrial revolution has made present-day atmosphere to be quite different from the natural atmosphere (Ashton 1948). Rapid industrialization and urbanization during this time led to increase in anthropogenic emissions from both fossil fuel and biomass combustion (Chowdhury et al. 2001). Particulate matter refers to the solid and liquid particles that are dispersed into ambient air. Particulate matter is composed of a mixture of particles directly emitted into the air (primary particles) and particles formed in the air from the chemical transformation of gaseous pollutants (secondary particles). There are two major categories of particulate matter (PM): PM_{10} and $PM_{2.5}$ (Martinez-Ramirez and Thompson 1998). The principal types of directly emitted particles are soil-related particles and organic and elemental carbon particles from the combustion of fossil fuels and biomass materials. The concentration of primary particles depends upon their

emission rate, transport and dispersion and removal rate from the atmosphere. Situation with secondary particles is much more complex (US EPA 1996). As a result, it is considerably more difficult to relate ambient concentrations of secondary particles to sources of precursor emissions than it is to identify the sources of primary particles.

Heavy metals are metallic elements which are toxic and have high densities, specific gravities or atomic weights (Duffus 2002). They constitute important class of pollutants which have received the attention of researchers all over the world, mainly due to their harmful effects on living beings. Heavy metals enter into the environment mainly via three routes: deposition of atmospheric particulates; disposal of metal enriched sewage sludge and sewage effluents; and by-products from metal mining processes (Shrivastav 2001). Metals in the atmosphere result from processes both natural (dust re-suspension, foliage emissions, volcanic activity and bubble bursting from water bodies) and anthropogenic (primarily industrial and automotive emissions; Harris 1976; Pacyna 1986). Re-suspension of roadside dust and soil is another potential source of heavy metals. Iron is a metal present in significant concentration in most emission sources of air pollution particles while sources of lead were linked to vehicles, re-suspended soil and oil burning (Schroeder et al. 1987).

Concentration range of particulate matter and atmospheric pollutant sizes

Schroeder et al. (1987) had reported 30–35 μgm^{-3} heavy metals in atmospheric particulate matter. Manganese, copper, zinc, cadmium, chromium, iron, nickel, potassium, calcium, vanadium, barium, arsenic, selenium and strontium are the most commonly found metals in the pollution sources and have been studied widely. In Western Europe, North America and Western Pacific, except China, annual mean total suspended particulates (TSP) concentrations range between 20 and 80 μgm^{-3} (Sivertsen 2002), and PM_{10} levels are between 10 and 55 μgm^{-3}. High TSP and PM_{10} annual mean concentrations are found in Southeast Asia (Sivertsen 2002) ranging between 100 and 400 μgm^{-3} for TSP and 100–300 μgm^{-3} for PM_{10}. High annual TSP concentrations of 300–500 μgm^{-3} are observed in the large cities of China. In Lahore (Pakistan), TSP mean annual values were 607–678 μgm^{-3} (Smith et al. 1996). Similarly, the PM_{10} levels in Indian cities have been found to be in the range of 100–400 μgm^{-3} (Sharma et al. 2003).

Effects of particulate matter

Many epidemiological studies have found a positive correlation between PM_{10} mass and respiratory illnesses and cardiovascular diseases (Dockery et al. 1993; Abbey et al. 1999). Other studies suggest that it is the finer particles, which have the greatest impact on health (Schwartz et al. 1996; Cifuentes et al. 2000), which is supported by the fact that $PM_{2.5}$ penetrates the human respiratory system more efficiently into the alveolar region. Nevertheless, studies have also found that higher concentrations of coarse particles ($PM_{2.5-10}$) can also increase overall mortality rates (Castillejos et al. 2000). Miguel et al. (1999) suggested that re-suspended road dusts contain materials which can potentially initiate allergic reactions. Studies on both rodents and humans demonstrate that ultrafine particles have a greater effect per unit mass than larger particles (Oberdorster 2001; Donaldson et al. 2000), while another study on asthma patients performed in Germany and Finland (Wichmann and Peters 2000) suggested that both fine and ultrafine particles have health effects which might be independent of each other. Some studies have also shown that the chemical composition of the particles may mediate the health effects.

Atmospheric transport and deposition of pollutants over long distances had received much attention, particularly in connection with the acid rain problem, the formation of photochemical oxidants and ozone, and the global climatic effects (National Academy of Science 1978). Together with wet deposition, particle dry deposition is responsible for delivering to ecosystems atmospheric loads of compounds such as SO_4^{2-}, NO_3^- and NH_4^+, base cations (such as Na^+, K^+, Ca^{2+} and Mg^{2+}) and heavy metals (e.g., Pb, Cd and Zn). Deposition of particles containing SO_4^{2-}, NO_3^- and NH_4^+ contributes to potential acidification and eutrophication of ecosystems. Base cation deposition can be important for nutrient cycling in solids, and it can also neutralize acid input. Heavy metals are known for their possible toxic effects (Schwartz and Slinn 1992). Measurement of heavy metals in particulate matter is, therefore, of immense importance for toxicological, environmental and occupational health studies (Pandey et al. 1998).

Air quality management and pollution control of particulate matter

Use of source apportionment method

For the effective management of air quality, great importance is attached to the identification of the sources of suspended particulates (Baek et al. 1997). Source apportionment can be achieved by using a variety of air quality models. Among these, receptor-oriented modeling is one of the conventional techniques and has been in broad use for source apportionment studies in the past decades (Ke et al. 2008). The framework for using receptor models to solve air quality problems consists of: formulating a conceptual

model; identifying potential sources; characterizing source emissions; obtaining and analyzing ambient particulate matter samples for major components and source markers; confirming source types with multivariate receptor models; quantifying source contributions with the chemical mass balance; estimating profile changes and the limiting precursor gases for secondary aerosols; and reconciling receptor modeling results with source models, emissions inventories and receptor data analyses (Watson et al. 2002).

Air quality surveillance method

Air quality surveillance is the procedure for assessing the concentrations of atmospheric contaminants and other properties of the air so that air quality management requirements can be met. Environmental regulators have access to a large set of policy tools and instruments to create incentives for industrial facilities to abate pollution. Command and control is one approach where pollution standards for emissions or effluence are set, and fines and penalties are assessed against non-compliant facilities. Other options include pollution charges, tradable permits, voluntary participation programs, as well as environmental performance rating and public disclosure programs (World Bank 2000).

Use of geographic information system (GIS) and global positioning system (GPS)

Other identified means of managing air quality include geographic information systems (GIS) and global positioning system (GPS). GIS are systems of hardware, software, data, people, organizations and institutional arrangements for collecting, storing, analyzing and disseminating information about areas of the earth (Dueker and Kjerne 1989). GIS helps in the assessment of quality of life (QOL) which is emerging as a major indicator to monitor citizen's livelihood and well-being at the grassroots level and thus inform local people and organizations of their living environment and optimize the allocations of resources to improve the community development (Hein et al. 2006; Loh et al. 2012). Global positioning system (GPS) receivers are another invaluable useful tool that can be used by state and local government agencies to address spatial coordinate deficiencies of inventory data of point sources (Alexis 2002). It consists of satellite, control and receiver segments in which the GPS receivers can be used to obtain emission release point locations if site access is available.

Remote sensing technique

Remote sensing is another technique of immense application in environmental pollution and involves collecting information about the earth without taking a physical sample of the earth's surface or touching the surface using sensors placed on a platform at a distance from it (Liu et al. 1996). A sensor is used to measure the energy reflected from the earth and information gotten can be displayed as a digital image or as a photograph. It was based on the fact that the atmosphere affects satellite images of the earth's surface in the solar spectrum and signal observed by the satellite sensor was the sum of the effects from the ground and atmosphere (King et al. 1999).

Meteorological and air pollution dispersion models

There are many meteorological models available for weather prediction and the mainly used are Weather Research and Forecasting (WRF) modeling system, MM5 modeling system software and Regional Atmospheric Modeling System (RAMS). MM5 is a limited-area, non-hydrostatic, terrain-following sigma-coordinate model designed to simulate or predict mesoscale atmospheric circulation (Grell et al. 1995). RAMS is a highly versatile numerical code developed by scientists at Colorado State University for simulating and forecasting meteorological phenomena, and for depicting the results (http://rams.atmos.colostate.edu/rams-description.html). Other applicable air pollution dispersion models for atmospheric air pollution assessment include photochemical modeling, plume rise models, particle models, deposition modules, odor models and statistical models (Yerramilli et al. 2011).

Previous studies on collection and assessment of particulate matter and heavy metals

Generally, sources of metals and particulate matter in the atmosphere had been traced to originate either from anthropogenic activities or natural phenomena such as dust re-suspension, foliage emissions, volcanic eruption and bubble bursting from water bodies (Harris 1976). However, different methodologies had been proposed in collecting samples of heavy metals and particulate matter; and assessing their quantities in the atmosphere.

Collection methods and measurement techniques

Lindberg and Turne (1988) had been reported to have used automatic rain sensing collectors, made of chemically inert materials, to collect wet-only samples of precipitation. Abdul-Wahab (2004) used nuclepore polycarbonate filters to collect atmospheric aerosol particles coupled with high-volume sampling method. Replicate inert surfaces situated in automatic collectors designed to exclude rainfall had been used to collect dry deposited particles (Lindberg and Lovett 1985). Lovett and Lindberg (1984) estimated dry

deposition of metal containing particles from inert surface fluxes using an empirical model of coarse particle dry deposition to the full canopy developed from measurements of throughfall chemistry. Many researchers had measured concentrations of heavy metals in particulate matter using graphite furnace atomic absorption spectrophotometry using the method of standard additions (Momani et al. 2000; Greaney 2005; Vijayananda et al. 2008; Farahmandkia et al. 2010; Awan et al. 2011; Dubey et al. 2012; Kleckerová and Docekalová 2014). In a different approach, Rojas et al. (1991) used a mathematical approach based on the Monin–Obukhov similarity theory to predict wind speed, friction velocity and drag coefficient which were then introduced into a well-known deposition model of Slinn and Slinn (1980) to calculate dry deposition of heavy metals into North Sea.

Elemental composition of particulate matter had also been determined using particle-induced X-ray emission (PIXE) method (Makra et al. 1999; Abiye et al. 2014), atomic absorption spectrophotometer (Sharma et al. 2003), benzene-soluble organic fraction method (Sharma et al. 2003), wet sieving method (Greaney 2005) and inductively coupled plasma quadrupole mass spectrometer (Samontha et al. 2007). Determination of particle number concentration using condensation particle counter (Marple et al. 1987), particle size distribution using electrical aerosol spectrometer (Mirme et al. 2002) and absorption coefficient using filter reflectance (Mathys et al. 2001) in particulate matter had been investigated (Vallius 2005). In another development, results obtained from an organic speciation analysis had been used in a chemical mass balance (CMB) model, along with organic emissions profiles and key organic tracers typical of urban sources, to quantify the primary source contributions to $PM_{2.5}$ mass concentrations in four major cities of India (Chowdhury et al. 2001). In a related work, Khodeir et al. (2012) modeled chemical composition data using factor analysis with Varimax orthogonal rotation to determine five and four particle source categories contributing significant amount of $PM_{2.5}$ and PM_{10} mass, respectively.

Statistical multivariate method and deterministic method had been applied as source apportionate methods for determining particulate matter sources whose chemical composition is stable and well defined (Vallius 2005). Samontha et al. (2007) used continuous-flow sequential extraction procedure to study fractionation and elemental association of some heavy metals in air particulate matter from two different sources: a smelter and heavy-vehicle traffic. In a new development, comprehensive comparison of positive matrix factorization (PMF) and molecular marker-based chemical mass balance (CMB-MM) modeling on $PM_{2.5}$ source contributions was conducted for particulate matter measurements taken at Jefferson Street, Atlanta, Georgia (Ke et al. 2008). Gaussian predictive model had been applied for model prediction of particulate dispersion from a cement mill stack (Otaru et al. 2013). Holnicki and Nahorski (2013) used Gaussian puff dispersion and regional-scale transport models for air quality modeling of Warsaw Metropolitan Area, Poland. In recent time, Xu et al. (2014) utilized a remote sensing method to retrieve PM_{10} concentration during health risk assessment of inhalable particulate matter in Beijing based on thermal environment.

Previous findings on particulate matter assessment

In the result obtained by Momani et al. (2000), atmospheric concentrations and enrichment coefficients of Zn, Cu, Pb and Cd were 344, 170, 291 and 3.8 ng/m^3; and 12.1, 6.1, 11.7 and 1.1, respectively. Various results obtained by different researchers revealed that concentration of heavy metals in particulate matter depends on the nature of the environment being examined. In marine environment, chlorine had been reported to be of higher concentration (Makra et al. 1999). Higher concentrations of sulfur and lead were traced to be partly of anthropogenic and biogenic origins. In another work involving source characterization of atmospheric heavy metals in industrial/residential areas (Abdul-Wahab 2004), copper and manganese were sourced from copper industrial processes and reinforcement steel production while chromium, lead and zinc reflected contributions from black and galvanized iron pipes production, mechanical industries, vehicle construction and motor-vehicle tire wear with municipal incineration. On the other hand, industrial site was polluting air quality in the examined residential area with copper, chromium, lead, nickel and manganese.

Sharma et al. (2003) had shown that heavy metals in Kanpur, India, were almost 5–10 times higher than levels in European cities with reference to $PM_{2.5}$ and PM_{10}. Diesel exhaust, gasoline exhaust, road dust, coal combustion and biomass combustion had also been identified as major sources of fine particle-phase inorganic and organic compounds in India (Chowdhury et al. 2001). In investigating fractionation and elemental association of metals in air particulate matter from smelter and heavy traffic sites sources, reducible phase (Fe/Mn oxides) was shown to be the largest fraction of Pb in air particulate matter from both the smelter and heavy traffic sites (Samontha et al. 2007). In the reducible fraction, Pb was found to dissolve earlier than Fe indicating that Pb was adsorbed onto Fe oxide surfaces in the air particulate matter. Ke et al. (2008) resolved four similar sources of $PM_{2.5}$ using positive matrix factorization (PMF) and molecular marker-based chemical mass balance (CMB-MM) modeling methods, with good correlation for road dust, but fair for gasoline

exhaust and wood combustion. The CMB-MM diesel exhaust has very poor correlation with the PMF resolved diesel exhaust. Heavy oil combustion, mixed industrial activities and traffic had been identified as major sources of heavy metals in $PM_{2.5}$ and PM_{10} (Khodeir et al. 2012).

The summary of previous studies executed on atmospheric assessment of particulate matter and heavy metals in the past four decades is presented in Table 1.

Mathematical modeling

The modeling equations include source apportionment, deposition rate prediction modeling and health risk characterization of PM_{10}. The source apportionment discusses enrichment factor analysis, chemical mass balance modeling, positive matrix factorization and factor analysis.

Source apportionment

Enrichment factor analysis

This is used for the comparison between the concentrations of atmospheric aerosol elements, when they are found to be higher than expected in their natural forms according to their proportions in the background aerosol. The enrichment factor is given as (Rahn 1976):

$$\mathrm{EF} = \frac{[C]_{\mathrm{sample}} \big/ [C_{\mathrm{ref}}]_{\mathrm{sample}}}{[C]_{\mathrm{crust}} \big/ [C_{\mathrm{ref}}]_{\mathrm{crust}}}, \tag{1}$$

where $[C]_{\mathrm{sample}}$ = concentration of C (analyte element), $[C_{\mathrm{ref}}]_{\mathrm{sample}}$ = concentration of the normalization reference element in the sample, $[C]_{\mathrm{crust}}$ = concentration in the continental crust, $[C_{\mathrm{ref}}]_{\mathrm{crust}}$ = concentration in the continental crust for the reference element.

Chemical mass balance modeling

The CMB receptor method tends to extract information about a source's contribution on the basis of the variability of elements measured in a large number of samples. The objective of this method is to detect common variability and imply source identity by comparing elements with common variability with elements associated with specific sources (Henry et al. 1984). At a given receptor site, a mass balance equation accounting for all conserved chemical species from p independent sources in a given sample can be expressed as follows (Ke et al. 2008; Jeong et al. 2011):

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij}, \tag{2}$$

where x_{ij} = concentration of the *jth* species measured in the *ith* sample, g_{ik} = mass contribution from the *kth* source to the *ith* sample, f_{kj} = fractional concentration of the *jth* species in the *kth* source, e_{ij} = residual associated with the ambient concentration of the *jth* species in the *ith* sample, p = total number of the contributed sources.

Positive matrix factorization (PMF)

Positive matrix factorization is one of the multivariate receptor models that utilize statistical techniques to reduce the data to meaningful terms for identifying sources of air pollutants and to estimate the source contributions (Qin and Oduyemi 2003). Its goal is to resolve the mixture of sources that contributes to particulate matter samples. Specifically, the mathematical model in matrix form is (Reff et al. 2007):

$$X = G \cdot F + E. \tag{3}$$

The index notation is as stated as Eq. 2. The goal is to find values of g_{ik}, f_{kj} and p that best reproduce x_{ij}. The values of g_{ik} and f_{kj} are adjusted until a minimum value of Q for a given p is found. Q is defined as:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{\sigma_{ij}} \right)^2, \tag{4}$$

where σ_{ij} = uncertainty of the jth species concentration in sample i, n = number of samples and m = number of species.

Various formulas of calculating uncertainties (σ_{ij}) for PMF analyses of PM data from previous literatures are presented in Table 2.

Factor analysis

Factor analysis is another multivariate receptor model in which data are transformed into a standardized form by normalizing the concentration of each element in each sample with respect to the mean value and standard deviation for the element. It begins with an eigenvector analysis of a cross product matrix of the data, frequently the matrix of correlation coefficients. Assuming each variable is linearly related to some number of underlying factors so that the values of variables may be expressed as a set of n linear equations, the factor model is given as (Gurjar et al. 2010):

$$X_{ij} = \sum_{k=1}^{p} C_{ik} S_{kj} + d_j U_{ij}, \quad k = 1, \ldots, m, \tag{5}$$

where

Table 1 Summary of previous studies on particulate matter assessment

References	Accomplished task	Research location	Methodology	Assessed PM and heavy metals	Conclusion
Lindberg and Turne (1988)	Factors influencing atmospheric deposition, stream export and landscape accumulation of trace metals in forested watersheds	Southeastern United States	1. Automatic rain sensing collectors 2. Replicate inert surfaces 3. Graphite furnace atomic absorption spectrophotometry	Cd, Mn, Pb, Zn and Al	1. Metal export was controlled by stream pH, organic carbon, bedrock geology and hydrologic characteristics of each site
Ruijgrok et al. (1995)	Assessment of current knowledge about dry deposition of particles with a focus on implications for Europe	Europe	–	–	1. Insufficient knowledge on particle dry deposition undermining reliable estimates of deposition fluxes over Europe
Shrivastav (2001)	Atmospheric heavy metal pollution: development of chronological records and geochemical monitoring	–	–	–	1. The problem of atmospheric heavy metal pollution would remain as a legacy of mass industrial activity for many generations and is likely to escalate further in future
Watson et al. (2002)	Receptor modeling application framework for particle source apportionment	USA	Enrichment factor, chemical mass balance, multiple linear regression, eigenvector, edge detection, neural network, aerosol evolution and aerosol equilibrium models	TSP, PM_{10} and $PM_{2.5}$	1. Compliance monitoring networks were not designed to obtain data for the observables, locations and time periods that allowed receptor models to be applied 2. Measurements from existing networks could be used to form conceptual models that allowed the needed monitoring network to be optimized
Abdul-Wahab (2004)	Source characterization of atmospheric heavy metals in industrial/residential areas	Oman	1. High-volume sampling method	Cr, Cu, Mn, Ni, Pb, Zn, Cd, V and Mo	1. Sources were linked to copper industrial processes and reinforcement steel production; black and galvanized iron pipes production; mechanical industries and vehicle construction; and motor-vehicle tire wear and municipal incineration

Table 1 continued

References	Accomplished task	Research location	Methodology	Assessed PM and heavy metals	Conclusion
Samontha et al. (2007)	Determination of fractionation of metals in air particulate matter from two different sources, a smelter and heavy-vehicle traffic	Thailand	1. Continuous-flow sequential extraction system 2. Extractograms	Cd, Zn and Pb	1. Crustal enrichment factors suggested that Cd, Zn and Pb in the air particulate matter were predominantly of anthropogenic origin 2. Reducible phase (Fe/Mn oxides) was the largest fraction of Pb in air particulate matter from both the smelter and heavy traffic sites 3. Total Pb deposition in the dry season was found to be higher than that in the wet season and may be attributed to soil dust 4. Pb was found to dissolve earlier than Fe indicating that Pb could occur adsorbed onto Fe oxide surfaces in the air particulate matter
Razos and Christides (2010)	Presentation of a different analytical methodology for samples of airborne particulate matter	Industrial area of Eleusis, Greece	1. Anodic stripping voltammetry (ASV) 2. Inductively coupled plasma atomic emission spectrometry (ICP-AES)	Zn, Pb, Cd, Cu, Fe, Mn, Ni, PM_{10} and $PM_{2.5}$	1. Correlation analysis between airborne particulate matter (PM) and toxic elements gave the sources that affected the presence of these elements in coarse and fine particulates
Lee and Hieu (2011)	Seasonal variations in mass concentrations of particulate matter and compositions of heavy metals in $PM_{2.5}$ and PM_{10} collected from a typical urban residential area	Ulsan, Korea	1. Inductively coupled plasma atomic emission spectrometry (ICP-AES)	$PM_{2.5}$, PM_{10}, Cd, Zn, Pb and Cu	1. Analysis of enrichment factors of heavy metals showed highly enriched Cd, Zn, Pb and Cu 2. Principal component analysis for heavy metals in $PM_{2.5}$ and PM_{10} identified industrial emissions and road dust (soil and traffic) as major sources at the sampling site
Khodeir et al. (2012)	Source apportionment and elemental composition of $PM_{2.5}$ and PM_{10}	Jeddah City, Saudi Arabia	1. Energy-dispersive X-ray fluorescence (ED–XRF) 2. Factor analysis with Varimax orthogonal rotation	$PM_{2.5}$, PM_{10}, Na, Mg, Al, Si, K, Ca, Ti, Cr, Mn, Fe, Rb, Sr P, V, Cr, Mn, Ni, Cu and Ba	1. $PM_{2.5}$ and PM_{10} sources were attributed to heavy oil combustion, re-suspended soil, industrial and traffic characterized by high concentrations of different heavy metals

Table 1 continued

References	Accomplished task	Research location	Methodology	Assessed PM and heavy metals	Conclusion
Otaru et al. (2013)	Model prediction of particulate dispersion from a Cement Mill Stack	Nigeria	1. Gaussian predictive model 2. SKC deployable particulate sampler 3. In-stack iso-kinetic sampling method 4. Vayubodhan stack sampler	PM_{10}	1. Simulated results were in agreement with experimental results at an average value of 92% within a Gaussian distance of 200–2000 m 2. Simulated safety distance beyond 7.0 km from the plant was recommended for human settlement and activities
Schröder et al. (2013)	Correlation between atmospheric deposition of Cd, Hg and Pb and their concentrations in mosses specified for ecological land classes covering Europe	Europe	1. Spatial autocorrelations	Cd, Hg and Pb	1. Spatial patterns of correlations between the atmospheric deposition and bioaccumulation were shown to vary by element and ecologically defined land classes
Holnicki and Nahorski (2013)	Air quality modeling	Warsaw metropolitan area, Poland	1. Gaussian puff dispersion model 2. Regional-scale transport model	–	1. Relatively low uncertainty $PM_{2.5}$ applied to forecasts of SO_2, while very substantial uncertainties related to PM_{10} and NO_x forecasts, which all strongly depended on very uncertain linear sources 2. Kind of polluting compound and emission source category were two main factors indicating the relationship of air pollution forecast
Habeebullah (2014)	Modeling particulate matter (PM_{10}) in Makkah, Saudi Arabia—a view point of health impact	Makkah, Saudi Arabia	1. Continuous IP beta gage monitor device 2. Air-Q 2.2.3 model	PM_{10}	1. PM_{10} levels in Makkah exceeded national and international air quality standards set for the protection of human health 2. This study was the first to apply Air-Q 2.2.3 model to provide quantitative data on the impact of particulate matter exposure on the health of people living in Makkah City, KSA, during 2012 (1433H)

Table 1 continued

References	Accomplished task	Research location	Methodology	Assessed PM and heavy metals	Conclusion
Abiye et al. (2014)	Receptor modeling of atmospheric aerosols	Federal Capital Territory, Nigeria	1. Sequential double-stage sampler 2. Positive matrix factorization (PMF) receptor model 3. Ion beam analysis (IBA) technique 4. Particle-induced X-ray emission (PIXE) technique	$PM_{2.5-10}$, Si, P, S, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, Cd, Sn, Ta and Pb	1. Three distinct sources, crustal 4.2%, biomass/fuel burning 8.8%, vehicular 87.0%, were identified and apportioned by PMF model 2. Traffic-related pollution posed a great hazard to the atmospheric environment of Abuja and likewise on human receptors in the studied areas 3. There was good correlation between on site measured concentrations and model predicted concentrations of PM

Table 2 Formulas for calculating uncertainties (σ_{ij}) for PMF analyses of PM data (Reff et al. 2007)

PMF uncertainty (σ_{ij})	References
$k_j \cdot x_{ij} + \frac{DL_{ij}}{3}$	Kim et al. (2005)
$(0.05 \cdot x_{ij}) + DL_{ij}$	Ito et al. (2004)
$\sqrt{(rep)^2 + (0.05 \cdot x_{ij})^2}$	Prendes et al. (1999)
$\sqrt{a_j s_{ij}^2 + b_j DL_{ij}^2}$	Polissar et al. (1996)
$0.3 + DL_{ij}$	Xie et al. (1999)
$s_{ij} + C_3 \cdot \lvert x_{ij} \rvert$	Chueinta et al. (2000)
$\bar{s}_j + \frac{\overline{DL_j}}{3}$	Polissar et al. (1996)
$s_{ij} + \frac{DL_{ij}}{3}$	Kim et al. (2004)
$s_{ij} + 0.2 \cdot DL_{ij}$	Ramadan et al. (2000)
$\sqrt{3 \cdot (s_{ij})^2 + DL_{ij}^2}$	Song et al. (2001)

s_{ij} analytical uncertainty, DL_{ij} method detection limit, C_3 value between 0.1 and 0.2, *rep* reproducibility, k fraction developed for each species by analyzing uncertainty vs. concentration plots, a_j, b_j scaling factors, *overbar* average

$$X_{ij} = \frac{X_{ij} - \overline{X}_i}{\left(\overline{X}_i^2 - \overline{X}_i^2\right)^{1/2}}, \tag{6}$$

$$S_{ik} = \frac{S_{kj} - \overline{S}_k}{\left(\overline{S}_k^2 - \overline{S}_k^2\right)^{1/2}}, \tag{7}$$

$$C_{ik} = C_{ik}\left(\frac{\overline{S}_k^2 - \overline{S}_k^2}{\overline{C}_i^2 - \overline{C}_i^2}\right), \tag{8}$$

where $X_{ij} = i$th elemental concentration measured in the jth sample, $\overline{X}_i$ = average of ith elemental concentration, S_{ik} = airborne mass concentration of material from the ith source contributing to the kth sample, S_{kj} = airborne mass concentration of material from the kth source contributing to the jth sample, $\overline{S}_k$ = average of airborne mass concentration of material from the kth source C_{ik} = gravimetric concentration of the ith element in material from the kth source, $\overline{C}_i$ = average gravimetric concentration of the ith element in material.

Deposition rate prediction modeling

Three major transfer processes are involved in the transportation of pollutants in the atmosphere at the most fundamental physical and chemical level (Brankov et al. 1998). These include convection, diffusion and transformation of air pollutants. This is represented by a system of partial differential equations, known as the Fickian system, representing the mass balance of each pollutant species in any elementary volume $dv = dxdydz$ in which the pollutant species appears. The basic equation is thus (Holnicki 2006; Jacobson 2005; Sportisse 2007):

[Accumulation of pollutant p in dv during time dt]
= [Rate of pollutants inflow into the element of volume]
− [Rate of pollutants outflow from the element of volume]
+ [Emission ratefrom pollution sources located in dv]
+ [Production anddestruction rates of the pollutant by chemical reaction]
− [Rate of pollutant removal through natural processes]. (9)

Mathematically,

$$\frac{\partial C_p}{\partial t} = -\nabla\left(uC_p\right) - \nabla q_p + E_p + R_p - S_p, \quad (10)$$

where C_p = concentration of pollutant p in volume dv, t = time (sec), u = velocity, q_p = mass flux of pollutant p due to turbulent diffusion, E_p = emission rate from pollution sources located in dv, R_p = production and destruction rates of the pollutant by chemical reaction, S_p = rate of pollutant removal through natural processes.

Equation (10) holds under the following assumptions:

- Rate of mass transfer due to diffusion is proportional to the concentration gradient.
- The chemical production rate involves the production of pollutant through the chemical interactions of other pollutants.

Under steady condition, Eq. (10) becomes

$$-\nabla\left(uC_p\right) - \nabla q_p + E_p + R_p - S_p = 0. \quad (11)$$

Health risk assessment of PM_{10}

A relative risk model based on Poisson regression, commonly used in epidemiological studies of air pollution to calculate the relative risk of inhalable particulates with certain health impacts, can be used to model PM_{10} health risk characterization (Kan and Chen 2004; Yaduma et al. 2013). Average relative risk of all health impacts can then be used to represent health risk of inhalable particulate matter using the following equations (Xu et al. 2014):

$$\mathrm{TR}_i = \frac{R_i}{R_{oi}} = e^{\beta_i \times (C - C_o)} \quad (12)$$

$$\mathrm{TR} = \frac{1}{n}\sum_{i=1}^{n} \mathrm{TR}_i, \quad (13)$$

where TR = health risk of inhalable particulate matter; TR_i = relative risk caused by the ith health impact; i = 1,2,3,… ,n; R_i = actual risk of the ith health impact; R_{oi} = reference risk value of the ith health impact; β_i = exposure − response coefficient; C = actual concentration of inhalable particulate matter, μg/m^3; C_o = -reference concentration in the risk assessment based on the average year guiding value of inhalable particulate matter set by the WHO, μg/m^3; n = number of health impact types caused by inhalable particulate matter.

Conclusion

1. Numerous literatures had presented applicable mathematical models to quantify the amount of particulate matter and heavy metals in the atmosphere; and prediction of their concentrations over time in the atmosphere. However, there is need for improvement on the mathematical models by reducing the number of assumptions made in developing the models such as
   - assuming linear relationship between variables of factor analysis model in order to have a set of n linear equations.
   - each air pollutant source type having unique chemical properties. Thus, highly correlated chemical compounds originate from the same source for multivariate receptor models such as positive matrix factorization.
   - using average relative risk of all health impacts to represent the health risk of inhalable particulate matter for health risk characterization of PM_{10} models.
   - using convection, diffusion and transformation of air pollutants only as transfer processes in the development of deposition rate prediction modeling.

   Also, fewer researchers presented the simulation of these models using existing experimental data. This was as a result of difficulties normally experienced in the course of gathering data as observed in some literatures. Thus, compliance monitoring networks should be designed to obtain data for the observables, locations and time periods that allowed receptor models to be applied. Also, measurements from existing networks can be used to form conceptual models that allowed the needed monitoring network to be optimized as suggested by Watson et al. (2002). There is need for further work on development of applicable computer software that will allow easy simulation of these models. This will enhance prediction of concentrations of these heavy metals, control and monitoring in the atmosphere.
2. In the examined literatures, various methodologies for assessing concentrations of heavy metals in the atmosphere have been presented. This enabled the determination of increasing order of heavy metal concentrations in the atmosphere at various locations

under experimentation. This has been linked to the heaviness of source apportionment of each heavy metal. However, there is need for further research on comparative analysis of concentrations of these heavy metals in the atmosphere under different methodologies. This will enhance the determination of accuracies of each methodology, and thus, correlation models can be formed between examined methodologies. Though order of increasing concentrations of heavy metals may vary from one environment to another based on source apportionment, it was observed that there was no consistency in the increasing order of concentrations of heavy metals in consulted literatures.

3. Particle shape and size are critical factors controlling the extent to which particles can penetrate into the respiratory tract, how and where particles are deposited, and at what rate particles are cleared form respiratory tract (Hassan 2006). Air-Q 2.2.3 model had been currently identified as a new methodology which had found its application in provision of quantitative data on the impact of particulate matter exposure on the health of people (Shakour et al. 2011; Habeebullah 2014). Further research works are required on this new methodology to increase the number of cases of hospital admissions due to respiratory diseases that can be handled per year by this model for each concentration range of particulate matter and heavy metals.
4. It has been observed that heavy metals have severe effects on plants, animals, humans and ultimately on environment (Sardar et al. 2013). There is need to compare the concentrations of heavy metals in each of the aforementioned in a single environment for future research work. This will enable the determination of the most susceptible of plants, animals and humans to heavy metal exposure and at the same time the most that has affinity for inhaling or absorbing particulate matter and heavy metals.
5. In all the examined literatures, none gave consideration to economic effect and impact of assessing heavy metals and particulate matter in the atmosphere. The cost implication of assessing, monitoring and controlling these unfriendly substances should be examined, and hence, involvement of cost engineers will be of immense help. Comparisons can be made for cost implication of methodologies of assessing atmospheric heavy metals and particulate matter to determine the one with better result and low cost.
6. Lot has been done on source apportionment to determine origin of each heavy metal in particulate matter present in the atmosphere. However, there is need for further research works that enable optimal control and regulation of emission of these heavy metals into the atmosphere in order to reduce health effects of these inhalable substances. Also, collection of comprehensive chemical composition information on airborne particles and measurement of composition of emissions from major source categories together with the application of organic source tracers would improve confidence in existing source apportionment models (Greaney 2005).

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