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Pant, Pallavi; Harrison, Roy

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Critical Review of Receptor Modelling for Particulate Matter: A Case Study of India

Pallavi Pant and Roy M. Harrison[*1]

Division of Environmental Health & Risk Management,
School of Geography, Earth & Environmental Sciences
University of Birmingham, Edgbaston

Birmingham B15 2TT
United Kingdom

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[*] Corresponding Author: Tel: 00 44 121 414 3494; Fax: 00 44 121 414 3709;
Email: r.m.harrison@bham.ac.uk

[1] Also at: Department of Environmental Sciences / Center of Excellence in Environmental Studies, King Abdulaziz University, PO Box 80203, Jeddah, 21589, Saudi Arabia.

ABSTRACT

India is used as a case study in reviewing the application of receptor models for source apportionment. India has high concentrations of airborne particulate matter, and the application of effective abatement measures is a high priority, and demands confidence in the results of source apportionment studies. The many studies conducted are reviewed, and reveal a very wide range of conclusions, even for the same city. To some degree these divergences may be the result of using different sampling locations and/or seasons, but to a large extent differences probably arise from methodological weaknesses. The assignment of factors from multivariate receptor models to specific source categories is in many cases highly questionable as factors often include combinations of chemical constituents that are of low plausibility. This ambiguity in terms of presence of tracer elements may be the result of genuine collinearity of diverse sources, or more probably arises from methodological problems. Few studies have used either organic molecular markers or chemical mass balance (CMB) models, and there is a shortage of data on locally-derived emission source profiles, although recent work has begun to remedy this weakness. The conclusions include a number of recommendations for use in design of future studies.

Keywords: Receptor Model; PM; Source Apportionment; Chemical Mass Balance; Multivariate Models; India

INTRODUCTION

Air quality has been a cause of concern all over the world with the concentrations of criteria pollutants exceeding the standards at many places, particularly in developing countries. Particulate matter (PM) has been recognized as one of the key pollutants with a negative impact on human health, and a range of regulations have been introduced in order to control PM_{10} levels in urban areas with an increasing focus on $PM_{2.5}$ control. However, in order to design effective programmes and strategies for reduction of PM concentration in the ambient air, it is necessary to have information about the sources and their respective contributions. The term, source apportionment (SA) describes techniques used to quantify the contribution of different sources to atmospheric PM concentrations. There is a wide range of published literature on source apportionment using dispersion models and monitoring data (Laupsa et al., 2009, Colvile et al., 2003). However, in the Indian context, most of the source apportionment studies have been conducted using receptor models and hence, receptor models are the focus of this review. Receptor models form a subset of source apportionment techniques and apportion the pollutant concentrations based on the measured ambient air data and the knowledge about composition of the contributing sources (Henry et al., 1984). The key outputs are the percentage contributions of different sources to pollutant concentration. Such models are particularly helpful in cases where complete emissions inventories are not available (Hopke, 1991). Receptor models have been used for identification of sources and their respective contributions to airborne particulate matter across the world (Harrison et al., 1997; Kumar et al., 2001; Larsen and Baker, 2003; Begum et al., 2004; Lai et al., 2005; Song et al., 2006; Tsai and Chen, 2006; Chowdhury et al., 2007; Guo et al., 2009; Kong et al., 2010; Stone et al., 2010; Gu et al., 2011).

Receptor models can be divided into two broad categories: microscopic and chemical. Microscopic methods, including optical, scanning electron microscope (SEM) and automated SEM analyses are primarily based on the analysis of morphological features of many individual particles in the ambient air (Cooper and Watson, 1980). However, they are not very feasible for large-scale use since they do not produce quantitative results in most cases. Chemical methods, on the other hand, utilize the chemical composition of airborne particles for identification and apportionment of sources of PM in the atmosphere. A number of different models are included in this category such as enrichment factor analysis, times series analysis, Chemical Mass Balance (CMB) analysis, multivariate factor analysis (including Principal Component Analysis (PCA) and Positive Matrix Factorization (PMF)), UNMIX,species series analysis and Multilinear Engine (ME) analysis (Cooper and Watson, 1980; Henry et al., 1984; Hopke, 1991; Ramadan et al., 2003). Such methods use trace elements, elemental/ organic carbon and organic molecular markers for identification of sources and over time have become popular for SA analyses.

Since PM is composed of both inorganic (trace metals, cations and anions) and organic species, a range of source markers are used in receptor modelling studies. Traditionally, most studies were carried out using inorganic trace elements like Fe, Zn, Pb, Cr, Al and Ni. However, since many of the trace elements are emitted from a range of sources (e.g., Zn is emitted from tyre wear as well refuse burning), it was difficult to apportion the PM to sources with a high degree of confidence. Further, with the removal of elements like Pb and Br from gasoline, there has been a need to develop and use new markers. In the last two decades, research has focused on the identification and development of organic molecular markers for SA since they can be characteristic of sources, thus reducing the source ambiguity, and creating markers for sources which are difficult to be apportioned solely on the basis of inorganic tracers, e.g., levoglucosan for biomass burning (Harrison et al., 1996; 2003; Schauer et al., 1996; Robinson et al., 2006).

The CMB method requires *a priori* knowledge of the composition of all sources contributing to the airborne pollution, but not their emission rates. The measured air quality is assumed to be a linear sum of the contributions of the known sources, whose contributions are summed over each different sampling period to give the best match to the concentrations of the many chemical species measured in the atmosphere. In more recent studies, organic "molecular markers" which may be only minor constituents of emissions are measured, as these help to discriminate between similar sources (e.g. gasoline and diesel engines).

There is a suite of multivariate statistical methods based upon factor analysis, of which PMF has been developed specifically for the purpose of source apportionment of air quality data, and is the most commonly applied. The method requires no *a priori* knowledge of source composition, but any information on source emissions characteristics is helpful in discriminating between similar sources. The method requires a substantial number (at least 50) of separate air samples and works best with a large dataset in which the number of samples far exceeds the number of analytical variables. A a minimum variable to case ratio of 1:3 should be maintained in order to obtain accurate results (Thurston and Spengler, 1985).

For a clearer distinction, it is better to have short sampling times so that overlap of multiple point source contributions to a given sample is minimised. The samples are analysed for the chemical constituents, and those constituents from the same source have the same temporal variation, and if unique to that source are perfectly correlated. Typically, however, a given chemical constituent will have multiple sources and the program is able to view correlations in a multidimensional space and can generate chemical profiles of "factors" with a unique temporal profile characteristic of a source. Past knowledge of source chemical profiles is used to assign factors to sources, and typically identification of six or seven different sources is a good outcome. Before PMF became widely adopted, PCA was widely used for the same purpose, but is less refined than PMF. Input data plays an important role in the final results, and care has to be taken to ensure that this is of good quality and where possible uncertainties can be assigned to individual analytes.

The key differences between CMB and the methods based upon multivariate statistics are summarised in Table 1. Studies have been conducted to compare results from different models (Larsen and Baker, 2003; Ramadan et al., 2003; Shrivastava et al., 2007; Bullock et al., 2008; Lee et al., 2008; Viana et al., 2008b; Yatkin and Bayram, 2008; Callén et al., 2009; Tauler et al., 2009). Multicollinearity can affect the model estimates, particularly in cases where different sources have similar signatures, although multivariate models help to reduce that problem substantially (Henry et al., 1984; Thurston and Lioy, 1987). It has been reported that in cases where two different sources have similar signatures, it becomes difficult to distinguish between them and neither CMB nor multivariate models can distinguish between sources with similar signatures when additional information (for e.g., meteorology data) is missing (Henry et al., 1984).

Hybrid models such as target transformation factor analysis (TTFA) and the constrained physical receptor model (COPREM) have been designed to combine the features of CMB and factor analysis models with the aim of maximizing the advantages while minimizing the limitations of each model (Wahlin, 2003; Viana et al., 2008a). The Multilinear Engine (ME) program also allows the use of source composition data to constrain the model.

Larsen and Baker (2003) compared three different multivariate techniques- UNMIX, PCA/MLR and PMF for SA of ambient polyaromatic hydrocarbons (PAHs) in Baltimore.

Although they reported that PCA/MLR is unable to model extreme data effectively, they concluded that the overall source contributions compare well among the various models. They also reported that use of different techniques on the same data set could help in identification of missing sources, and increase the robustness of the results. Shrivastava et al. (2007) used PMF and CMB for source apportionment of organic carbon and found good correlation between individual profiles for CMB and factors identified by PMF but with systematic biases that were found to be within an acceptable range (a factor of two). Lee et al. (2008) compared the CMB and the PMF models and concluded that although both models identify similar sources, they apportion contributions of different sources differently. The authors suggested that a lack of local source profiles, omission of key sources or lack of suitable markers, and the different assumptions regarding aging of the source emissions as the possible causes for the different estimations. Viana et al. (2008b) compared PCA, CMB and PMF for identification of source contributions to PM_{10} in Spain. They reported overall consistency between the different models with high correlation in terms of source identification. However, they noted larger differences in terms of the percentage contribution of various sources. They suggested that a combined approach with the use of multivariate techniques for identification and interpretation of emissions sources and use of CMB for source contribution could help in increasing the robustness of the results. Earlier, Thurston and Lioy (1987) had also suggested a similar approach with the consecutive use of multivariate and chemical mass balance models to derive better results from receptor modelling studies. Similarly, Shi et al. (2011) tested a combined two-stage PCA/MLR- CMB model and found acceptable results using synthetic datasets with collinearity. They also concluded that maximum uncertainty is generally observed in case of highly collinear sources.

Callén et al. (2009) compared three different multivariate techniques- PCA-ACPS, UNMIX, PMF for source apportionment of PM_{10} and found that the different models showed high correlation between modelled and measured concentrations and PCA and PMF were able to identify more sources in comparison with UNMIX with good agreement. Tauler et al. (2009) compared four different multivariate models (PCA, PMF, Multivariate Curve Resolution by Alternating Least Squares, (MCR-ALS) and Weighted Alternating Least Squares (MCR-WALS)) and concluded that PMF and MCR-WALS identify sources and apportion the emissions to sources in a similar fashion. The weighted models (PMF and MRC-WALS) were found superior in robust and accurate factor identification.

Receptor models have been used for regulatory purposes since they were first used in Oregon, USA in the late 1970s (Gordon, 1988). However, there is a caveat regarding the degree of uncertainty associated with the results (Caselli et al., 2006).

Air Pollution in India

Given the rapid rates of urbanization in Indian cities, air pollution is increasingly becoming a critical threat to the environment and to the quality of life among the urban population in India. Air quality has been a cause of concern in Indian cities with the concentrations of criteria pollutants exceeding health-based standards, and PM has been identified as one of the key public health concerns. High enrichment factors have been reported for various metals including Pb, Zn, Cu, Ni and Cr in a number of Indian cities, indicative of anthropogenic sources of heavy metals in particulate matter (Kulshrestha et al., 1995; Pandey et al., 1998; Negi et al., 2002; Rastogi and Sarin, 2009). Also, using SEM-EDX analysis, Srivastava et al. (2009b) reported that particles were primarily of anthropogenic origin irrespective of size range in polluted areas, e.g. traffic intersections. Although there has been an increased focus on PM emission control in recent years, the concentrations are still found to exceed the National Ambient Air Quality Standards (NAAQS) regularly.

The primary sources of air pollution in India have been identified as vehicular emissions, industrial emissions, coal combustion, biomass burning, road dust and refuse burning. There has been a rapid increase in motorization in India in the past years and this has led to an increasing contribution of the transport sector to air pollution in urban areas. Small-scale industries functioning within urban centres have been found to contribute to the air pollution problem.

SOURCE APPORTIONMENT AND RECEPTOR MODELLING IN INDIA

There has not to our knowledge previously been a review of either aerosol source apportionment or receptor modelling work conducted in India. In this article, we seek to review existing knowledge and to make recommendations as to future directions. There is a growing body of literature on source apportionment of PM in India using receptor modelling (Table S1 in Supplementary Information). A majority of the SA studies have been conducted using multivariate methods; PCA being the most commonly used technique although there are some cases of application of the CMB model (Gupta et al., 2007; Srivastava et al., 2009a;

Gummeneni et al., 2011). One of the key reasons for use of multivariate models is the absence of local source profiles, and it is only in the recent times that source profiles have been generated for some of the sources in India. Most of the studies using CMB have used the source profiles available through the USEPA Speciate database. Gupta et al. (2007) prepared soil dust and road dust source profiles for Kolkata, and the recently released Central Pollution Control Board (CPCB) source apportionment study (CPCB, 2010) also contains India-specific source profiles for a number of sources. Use of microscopic methods has also been reported though it has not been widely applied (Negi et al., 1987; Bandhu et al., 2000; Srivastava et al, 2009b). Enrichment factor analysis has been used in several cases, either in conjunction with factor analysis or independently (Negi et al., 2002; Shridhar et al., 2010). One study has been reported using PMF analysis (Bhanuprasad et al., 2008) although it was conducted at a regional scale (an Indian Ocean cruise) rather than at the city level. There are only a couple of studies comparing results from different models, and in both cases, a comparison has been made between the CMB model and multivariate methods (Srivastava et al., 2008; Srivastava et al., 2009a). Results of both the studies have indicated overall similarity akin to other studies (Shrivastava et al., 2007; Lee et al., 2008).

Total suspended particulate matter (TSP) and PM_{10} have been studied in great detail due to concerns over the health impacts but in recent years, the smaller size fractions of particulate matter ($PM_{2.5}$ or less) have also gained immense importance given the recent evidence of their public health implications. However, much of the research is still focused on TSP and $PM_{10,}$ with the exception of a few cases where $PM_{2.5}$ (Chowdhury et al., 2007; Tiwari et al., 2009; Khare and Baruah, 2010; Gummeneni et al., 2011) or PM_1 have been analysed (Chakrobarty and Gupta, 2010).

In terms of geographic distribution across the country, most of the studies have been conducted in New Delhi (Balachandran et al., 2000; Khillare et al., 2004; Srivastava and Jain, 2007b; Shridhar et al., 2010) followed by Mumbai (Kumar et al., 2001; Chelani et al., 2008; Kothai et al., 2008) and Kolkata (Gupta et al., 2007; Kar et al., 2010). A few studies have been conducted in other cities including Kanpur (Chakrobarty and Gupta, 2010), Chandigarh (Bandhu et al., 2000), Agra (Kulshrestha et al., 2009; Masih et al., 2010), Tirupati (Mouli et al., 2006) and Hyderabad (Gummeneni et al., 2011). In most of the studies, a range of urban sampling sites have been used for analysis including residential, commercial, industrial, and traffic intersections/kerbside. However, there are a limited number of analyses focused on measurement and analysis between urban and background locations (Chowdhury et al., 2007; Shridhar et al., 2010) where results have indicated significant enrichment of trace metals in the urban environment. The Central Pollution Control Board report (CPCB, 2010) referred to in more detail later, is based upon detailed studies conducted in six individual cities.

A majority of the PM source apportionment studies have been conducted using trace element markers (Balachandran et al., 2000; Kumar et al., 2001; Srivastava and Jain, 2007b; Kothai et al., 2008) and in some cases, inorganic tracers have been used in conjunction with organic and elemental carbon (Gupta et al., 2007; Tiwari et al., 2009; Chelani et al., 2010). The use of organic molecular markers for PM source apportionment has only been reported in recent years (Chowdhury et al., 2007; Masih et al., 2010).

Source Profiles

One of the most important parameters in the receptor modelling analysis is the selection of source signature/source profile. Source signature refers to the mix of tracer elements and/or molecular markers used for identification of sources. A wide range of source signatures have been used by authors for source identification across the country (Table 2) and the key markers used in the SA studies conducted in India are more or less similar to the markers reported/used internationally. These include the use of Al, Si, Ti, Ca etc for crustal/soil sources, Ni and V for residual/fuel oil combustion, Zn, Cr for refuse burning/incineration and Zn, Cr, Pb, Cu etc for vehicular emissions (Refer to Table SI for details).

In terms of harmonizing source profiles for source identification and apportionment, one of the key considerations is the fact that different researchers have interpreted sources differently. Based on a review of the available literature, the following key source signature categories can be classified for emission sources in India:

Crustal re-suspension/soil dust/ road dust: These are often difficult to separate, particularly as road dust is often a complex mixture from various anthropogenic and biogenic sources including vehicular exhaust, coal combustion, soil dust, construction material and industrial emissions that contribute to its composition through dry deposition (Rogge et al., 1993). Crustal elements typically used as tracers for soil dust and/or crustal re-suspension include Al, Si, Ca, Ti, Mg, Fe and Na (Lough et al., 2005; Jaeckels et al., 2007; Begum et al., 2010; Stone et al., 2010; Yin et al., 2010; Fujiwara et al., 2011; Gu et al., 2011). A whole array of element tracers has been used in India for identification of this source type including Al, Si, Ca, Ti, Fe, Pb, Cu, Cr, Ni, Co, Mg, and V (Balachandran et al., 2000; Khillare et al., 2004; Chelani et al., 2008; Chakrobarty and Gupta, 2010; Shridhar et al., 2010; Srimuruganandam and Nagendra, 2011). Certain of these are rather surprising (eg Ni, Co and V) as their abundance in soil is low and in vehicles only modest.

Elements like scandium (Sc) or Fe are sometimes considered, based on the rock type of the area under study. For example, in the case of New Delhi, Fe has been used as a marker for crustal re-suspension by many authors since rock composed of ferrogenous quartzite from the Aravalli Hills is representative of the source material for soils within Delhi itself. Brake dust is also a source of Fe (Gietl et al, 2010), potentially leading to confusion of the two sources.

Different authors have interpreted the sources of soil/crustal dust, road dust and re-suspended dust differently, and it is difficult to compare results since the category types are highly varied. While some authors have reported soil dust and crustal re-suspension due to vehicular activity as separate sources (Gupta et al., 2007), others have combined the two (Kulshrestha et al., 2009). In some cases, construction dust has also been included in the crustal source category. A number of authors have used the assumption that soil dust includes loadings of metals like Pb, Cr, Ni, Co etc. from various sources due to deposition over time (Bandhu et al., 2000; Mouli et al., 2006; Srivastava et al., 2007; Chakrobarty and Gupta, 2010), while other have segregated soil dust and road/ re-suspended dust using the absence/presence of crustal elements like Ca, Si, Al and metals Zn, Pb and Cr in the source profile (Gupta et al., 2007; Chowdhury et al., 2007; Kothai et al., 2008; Kulshrestha et al., 2009; Khare and Baruah, 2010). In some cases, the source identification is ambiguous given the choice of markers used for the source. For example, Basha et al. (2010) have identified the factor containing Pb, Cr, Co as soil/ re-suspended dust, and factors containing Cu and Cd as road dust. Given that there are a number of plausible sources for Pb, Cu, Cr, and Co, it is difficult to ascertain if the factor actually corresponds to the soil source. Similarly, Mouli et al. (2006) reported a factor enriched in Mn, Ni, Co, Cu, Pb, Li, V and Cr as a crustal source with contamination from coal fly ash, and vehicular emissions.

The crustal/road dust source has been reported to be a major contributor in the case of Delhi, with distinct seasonal patterns, where as much as 70 percent of the coarse fraction of PM_{10} (PM between 2.5-10 µm) has been attributed to crustal re-suspension (Balachandran et al., 2000; Khillare et al., 2004; Srivastava and Jain, 2007b; Srivastava et al., 2009b; Chelani et al., 2010; Shridhar et al., 2010). A significant correlation has been reported between SPM mass and Al, Ca, and Mg for Delhi (Shridhar et al., 2010). Higher concentrations of the coarse fraction of particulate matter in the summer season in Delhi are attributed to the hot winds that blow across from the Thar Desert in Rajasthan (Khillare et al., 2004; Srivastava et al., 2009b). This source is also found to be contributing substantially in other cities including Chandigarh (Bandhu et al., 2000).

Gupta et al. (2007) prepared local source profiles for road dust and soil dust for Kolkata and concluded that 36% and 17% of TSP was caused due to soil dust and 16% and 17% due to road dust at industrial and residential sites respectively. Major components of road dust and soil dust were found to be OC, TC, Fe, SO_4^{2-} and OC, TC, Cl^- and SO_4^{2-} respectively. The presence of Cl^- and SO_4^2 at the residential site was attributed to a marine aerosol influence, but sea salt was not identified as a PM component by the CMB model.

Meena et al. (2011) used environmental magnetism for identification and characterization of polluted areas in New Delhi and found that soil in industrial and traffic areas contains high concentrations of heavy metals in the soil. This obviously leads to difficulties in discriminating soil-derived particles from industrial emissions and road dust.

Vehicular sources: Emissions arising from road vehicles are generally contributed by a mix of tailpipe emissions, and wear and tear of brakes and tyres and re-suspension of road dust (Rogge et al., 1993; Thorpe and Harrison, 2008). A series of markers, both elemental and organic, have been used for source attribution of PM to the vehicular sources including composite emissions, petrol and diesel emissions, tyre and brake wear, and crustal re-suspension due to the vehicular activity. According to international research, elemental markers include Cu, Zn, Pb, Ni, Mn, Mo and Sb (Lough et al., 2005; Almeida et al., 2006; Crawford et al., 2007; Begum et al., 2010) while molecular markers include hopanes and steranes (Rogge et al., 1993; Watson et al., 2002; Lough et al., 2005; Jaeckels et al., 2007; Yin et al., 2010; Fujiwara et al., 2011). However, since the introduction of unleaded petrol, the concentrations of Pb are seen to be reducing progressively (Stone et al., 2010), and hence, Pb is not used as a stand-alone marker for vehicular emissions. When gasoline is entirely lead-free, there are minor contributions to lead in air from brake pads and road dust re-suspension. Lower molecular weight n-alkanes (typically C_{19-25}) are also found in vehicular exhaust (Rogge et al., 1993) and barium (Ba) has also been used a tracer for brake and tyre wear emissions (Lough et al., 2005; Gietl et al., 2010). Crustal elements like Fe and Al have also been reported to be found in diesel exhaust by Wang et al. (2003) but contributions from this source are likely to be very small. It has been established that the lead in coarse dust is generally contributed by the road dust that is re-suspended due to vehicular motion and lead reprocessing facilities while lead in the fine mode comes primarily from leaded gasoline, and in some cases, from battery recycling plants (Santoso et al., 2011).

Internationally, elemental carbon (EC) (Song et al., 2006; Fujita et al., 2007; Lee et al., 2008; Robles et al., 2008; Stone et al., 2010; Yin et al., 2010) and chrysene, fluoranthene and pyrene (Larsen and Baker, 2003; Kleeman et al., 2008) are used extensively as markers for diesel exhaust while coronene, benzo(ghi)perylene and indeno(1,2,3-cd)pyrene are tracers for petrol vehicle exhaust (Fujita et al., 2007; Bullock et al., 2008; Kleeman et al., 2008). Pyrene, fluoranthene and phenanthrene have been reported to be present in tyre debris (Rogge et al., 1993). PAH are reported to be enriched in road dust and roadside soil (Smith et al., 1995; Agarwal, 2009), hence complicating the differentiation of this source from direct vehicle emissions.

In India, Pb is the most commonly used tracer element for identification of vehicular emissions; other elements including V, Mn, Co and Zn (Balachandran et al., 2000; Kumar et al., 2001; Chelani et al., 2008). Low-lead petrol was introduced in Delhi in 1994 followed by unleaded petrol in 1995. However, it was made mandatory only from 1998 and in the rest of the country from 2000. Lead is still used as a marker for traffic emissions as it is reported to be still present in the road dust/ soil due to its long residence time (Banerjee, 2003). However, a recent analysis by Vijayanand et al. (2008) found no significant correlation between Pb and SPM and this was attributed to the introduction of unleaded petrol, and the subsequent decrease in the Pb emissions. Studies in Delhi report lead concentrations of 0.66 $\mu g/m^3$ (Balachandran et al., 2000), $0.38 \pm 0.32\ \mu g/m^3$ (Khillare et al., 2004), $0.37 \pm 0.12\ \mu g/m^3$ to $1.04 \pm 0.20\ \mu g/m^3$ in the coarse fraction of PM_{10} and $0.22 \pm 0.12\ \mu g/m^3$ to $0.94 \pm 0.17\ \mu g/m^3$ in the fine fraction (Srivastava et al., 2009a) and 0.039 ± 0.005 to $0.087 \pm 0.014\ \mu g/m^3$ (Chelani et al., 2010). Platinum (Pt), palladium (Pd) and rhodium (Rh), components of automotive catalytic converters, were found to be significantly correlated in road dust samples, and higher concentrations were reported for samples from road junctions and traffic signals (Mathur et al., 2011). Similar results have been reported by Lough et al. (2005). Of the elements reported as tracers of vehicular emissions (Table 2) the inclusion of Cr and Co seems hard to justify – industrial emissions are more likely sources of these elements. Fe, Mn, and Cu, and to a lesser extent Pb are present in brake wear particles (Thorpe and Harrison, 2008; Gietl et al., 2010) and zinc is enriched in tyre wear particles. Ca is present in lubricating oils, and Harrison et al. (2003) reported enrichment in the nanoparticle fraction of vehicle exhaust. In PM_{10} (and to a lesser extent $PM_{2.5}$) concentrations of Ca are likely to be dominated by crustal sources.

EC, organic carbon (OC), hopanes and steranes have been used for differentiation between petrol and diesel vehicles in India (Chelani et al., 2010). Organic markers used include hopanes and steranes, and PAHs (Sharma et al., 2003; Chowdhury et al., 2007; Sharma et al., 2007; Masih et al., 2010). B(b)F, B(a)P, IP and B(ghi)P and B(a)A+ Chry, IP and Flan have been used as markers for petrol and diesel emissions respectively (Sharma et al., 2007). Vehicular emissions are a major source of PM and research indicates that they contribute between 10- 80 percent to particulate matter in cities across India. Comparison of such estimates is made difficult by the fact that the various studies have quantified different vehicular sources (exhausts, resuspension, abrasion, etc). Also, there has not been much detailed analysis regarding the source profiles for the different vehicle types typically found in India.

Biomass burning: Potassium (K) is used as a tracer of crustal dust in the coarse range and soluble K for biomass burning in the fine range of particulate matter. In India, it has been used a key elemental marker for biomass/wood combustion for TSP, PM_{10} and $PM_{2.5}$ (Khare and Baruah, 2010; Shridhar et al., 2010) while levoglucosan is the key organic marker (Chowdhury et al., 2007).

Potassium and NH_4^+ have also been used as markers for wood burning and agricultural activities (Khare and Baruah, 2010). In some cases, wood and biomass burning have been combined as one source, with K used as the marker.

Biomass combustion has been found to contribute in the range of 7-20 percent depending on season and location. It has been found to be one of the major sources in Delhi, particularly in winter due to the combustion of wood (Sharma et al., 2003; Chowdhury et al., 2007). However, in other cities, biomass combustion has not been found to contribute substantially to PM.

Industrial emissions: A range of tracers have been used for identification of industrial emissions including Cu, Cr, Mn, Ni, Co, Zn etc. Table 3 presents the sources of heavy metal as listed in the UK heavy metal emissions inventory. This will not, of course, be directly representative of India, but gives an idea of the main sources of commonly reported metals.

In international studies, Begum et al. (2004) used Ni, Pb and S as markers for industrial emissions, Song et al. (2006) used Ni, Cr, Fe and Mn, Yatkin and Bayram (2008) used Fe, Mn, Zn, and Pb, Lestari and Mauliadi (2009) used Zn and Fe as markers for the steel industry, and Al and Cr as markers for the electroplating industry and Tauler et al. (2009) used Zn, Fe, Mn and Cd as tracers of steel metallurgy.

A range of trace elements have been used as markers for industrial emissions in India. Negi et al. (1987) distinguished between textile industry emissions (V, Br), oil refinery emissions (S, Cu, Ni, and V) and non-ferrous industry emissions (Zn, Cu, and Mn). Similarly, Mouli et al. (2006) used Cd, As Li, V, and Cu as markers for lead acid/ non-ferrous industry, and Mo, Fe, Se, Sb as markers for metallurgy. Kar et al. (2010) have also distinguished between tannery (Cr), industry (Co), and electroplating/galvanizing units (Zn, Cu, Ni). Shridhar et al. (2010) distinguished between industrial emissions (Ni, Cr, Mn, Cu, Zn) and emissions from battery units (Pb). Pb can also be emitted from paints, alloys, and plastic and rubber industries (Haritash and Kaushik, 2007).

There is clearly immense overlap of the marker elements attributed to industry with those associated with vehicular emissions, refuse/solid waste burning, crustal/road/resuspended dust and construction activities in Table 2. Given the similarity in the markers used for vehicular and industrial emissions, some authors have used markers like Cd, Cr, Ni, Pb, Zn, Cu etc. and identified the source as vehicular and industrial emissions (Khillare et al., 2004; Kothai et al., 2008; Khare and Baruah, 2010). A significant correlation was found between concentrations of Pb, Cr, Ni and Cd in New Delhi (Khillare et al., 2004) suggesting a common source, or a common meteorological control of concentrations.

Refuse burning/incineration: Key markers include zinc (Zn), chromium (Cr), and nickel (Ni). However, it is difficult to compare the source attributions as some authors have combined refuse burning, incineration and hazardous waste disposal, while others have restricted the source to incineration or refuse burning. In such cases, it is difficult to ascertain the exact contribution of the various activities. Further, since waste management is often conducted in an ad-hoc manner in Indian cities, it is difficult to ascertain the exact nature as well as well as the contribution of such a source. Sharma et al., (2003) found that refuse burning contributed significantly to the organic fraction of PM. In US studies, Zn and Cr have been used as markers for refuse burning/ incineration by Schauer et al. (1996) and Bullock et al. (2008).

Coal combustion: In international studies, key markers for coal combustion include As, Se, Te and SO_4^{2-} and it has been found to contribute between 6-30 percent to particulate matter in different studies (Khare and Baruah, 2010; Kumar et al., 2001; Gupta et al., 2007; Sharma et al., 2007; Srivastava and Jain, 2007b). Selenium (Se) has been reported to be a good marker for coal combustion (Hien et al., 2001; Lee et al., 2008). Other markers include Al and Si (Bi et al., 2007; Zeng et al., 2010), Zn (Almeida et al., 2006) and Cl (Song et al., 2006). Use of Al and Si opens the possibility of confusion of coal fly ash with crustal dusts, and Zn with traffic and/or refuse burning emissions. The use of Cl depends upon conversion of gaseous HCl emissions to particulate NH_4Cl which is not favoured by the generally hot Indian climate (Pio and Harrison, 1987). Picene has also been used as a marker for coal combustion (Stone et al., 2010; Yin et al., 2010). In general, Indian coal has a low sulphur content in comparison with other coals (Bhanarkar et al., 2008; Chikkatur, 2008) although the sulphur content of coal is reported to be high in eastern India (3.6-4.4%) (Khare and Baruah, 2010). Stone et al. (2010) also reported the use of high-sulphur coal in India while Negi et al. (1987) also reported Pb and Zn to be present in increased concentrations due to the use of domestic soft coal. Also, Indian coal is reported to have high ash content (Khare and Baruah, 2010) which is likely to elevate concentrations of crustal elements.

For New Delhi, where three coal-fired power plants are sited within the city boundaries, Sharma et al. (2007) attributed ~ 17 percent of the variance as per the PCA results to coal combustion while Srivastava and Jain (2007b) attributed approximately 15 percent of the variance of the PM 0.7 fraction to the source. Gupta et al. (2007) attributed nearly 40 percent of the TSPM and PM_{10} to coal combustion for a residential site in Kolkata.

Marine aerosols: Key markers include sodium (Na), potassium (K), chloride (Cl^-), and magnesium (Mg). However, since most of the source apportionment studies have been conducted in inland cities, marine aerosols are not found to be a key source, except in the case of Mumbai, a city with a huge coastline. Use of K offers possible confusion with wood/biomass combustion and Cl with coal burning, but a combination of the four elements should provide a reliable signature.

Authors have attributed between 9 (Kothai et al., 2008) to 30 (Chelani et al., 2008) percent of particulate matter to marine aerosols.

Secondary aerosols: Key markers include NO_3^-, SO_4^{2-}, NH_4^+. There has been very limited analysis of secondary aerosols and their contribution to PM mass has not been reported widely. Chakrobarty and Gupta (2010) reported secondary aerosols to be contributing approximately 39% to the overall PM_1 mass. Khare and Baruah (2010) attributed 8% of the PM2.5 in the city to secondary aerosol formation wherein NH_4^+ was attributed to agricultural and industrial activities and SO_4^{2-} was attributed to coal combustion and wood burning. Sulphate has been used as a marker for coal combustion in some Indian studies whereas NH_4^+ has been used as a marker for biomass combustion (Refer to Table 2).

Other sources that have been identified to contribute to PM mass in Indian cities include building/ construction dust (Srivastava and Jain, 2007a,b; Shridhar et al., 2010); refuse oil burning (Shridhar et al., 2010), wind-blown dust (Shridhar et al., 2010), two-stroke emissions with fugitive dust (Kothai et al., 2008) and tyre wear (Gupta et al., 2007). Karar and Gupta (2007) attributed 8 % of the variance to cooking.

Source Apportionment of Particulate Matter in Delhi

Delhi, the capital city of India, figures among the most polluted cities and presently faces the twin challenge of managing the demands of a growing city while still maintaining the quality of the environment. Concentrations of air pollutants in Delhi are often found to exceed the NAAQS despite repeated efforts to reduce air pollution. Delhi is reported to have nearly 33 different industrial areas (http://industries.delhigovt.nic.in/functions/faq.html#) and there are a large number of small-scale industries including electroplating, pickling and galvanizing (Banerjee, 2003). Delhi also has three coal-fired power plants (Badarpur, Indraprastha and Rajghat) and the vehicle population is among the highest in the country. The sulphur content in the coal used in power plants in Delhi typically ranges between 0.35% and 0.50% (Chowdhury et al., 2007). Several studies have been undertaken for source apportionment of suspended particulate matter (SPM), PM_{10} and $PM_{2.5}$ using receptor modelling in Delhi and most of them have attributed a large percentage of the PM to vehicular emissions, road dust and coal combustion (Balachandran et al., 2000; Khillare et al., 2004; Chowdhury et al., 2007; Sharma et al., 2007; Srivastava et al., 2008; Chelani et al., 2010; Shridhar et al., 2010). Goyal et al. (2010) have reported diesel vehicles to contribute nearly 28 percent of the total particulate matter in Delhi. Several other sources have also been reported for PM emissions in Delhi including open refuse burning (Khillare et al., 2004) and domestic use of biofuels and/or fossil fuels including kerosene (Mönkönnen et al., 2004). Sharma et al. (2007) attributed an increase in the concentration of PAHs to the increase in vehicle number and concluded that PAH release is dominated by vehicular emissions across all seasons in Delhi.

Agarwal (2009) reported an average value of 4694 ± 3028 μg/kg for PAHs in soils at traffic sites while Sarkar et al. (2010) reported positive correlation between PAHs and markers for industrial and vehicular emissions including Zn, V, Ni, Cr and Cu.

Balachandran et al. (2000) conducted source apportionment for coarse and fine fractions of PM_{10} using inorganic markers and PCA and found vehicular sources, industrial sources and crustal re-suspension to be the key contributors with each of them corresponding to three factors that explained 53.9, 19.4 and 15.7 percent of the variance respectively. Similar results were obtained by Khillare et al. (2004). Sharma et al. (2003) analysed particle phase organic compounds and identified vehicular sources, biomass burning and/or refuse burning as the key sources for the organic fraction of the particulate matter. Chowdhury et al. (2007) conducted the source apportionment for $PM_{2.5}$ using organic markers in New Delhi, Kolkata, Mumbai and Chandigarh. In the case of Delhi, they identified emissions from fossil fuel combustion (coal, diesel, and petrol) as responsible for about 25–33% of $PM_{2.5}$ mass, and biomass combustion for about 7-20%. Srivastava and Jain (2007a) used CMB for source apportionment of particulate matter and found that vehicular sources contribute between 60-90% to the fine size PM while paved road dust, crustal re-suspension and solid waste etc contribute primarily to coarse particulate matter.

Tiwari et al. (2009) reported PM_{10} and $PM_{2.5}$ to be composed of undetermined fractions, secondary inorganic aerosols, salt aerosols and mineral matter. Chelani et al. (2010) found auto-exhaust and re-suspension of crustal dust to be the key contributors with industrial species, secondary aerosol and refuse burning contributing at specific sites. Shridhar et al. (2010) reported re-suspended dust, construction dust, industrial activities and biomass burning as the key sources of SPM and associated metals, and also noted that the total mass concentration of trace metals contributed circa 2% to the SPM at the urban site while at the rural site, it contributed less than 1%. They also found the SPM concentration at the urban site to be significantly correlated with Ca, Mg and Al. Contrary to most other source apportionment results for Delhi, Srivastava and Jain (2008) reported a minimal contribution of soil crustal dust to the SPM concentration.

Almost all of the studies in Delhi have used the factor analysis (PCA) method for source apportionment, and results are similar to the extent that re-suspended dust and vehicular emissions are found to be the major contributors. Other key sources include biomass combustion, solid waste burning, coal combustion and construction dust. Results from CMB analysis have also yielded similar conclusions. However, LPG combustion was found to be a major contributor in residential areas in the city according to the results of CMB analysis as reported in CPCB (2010).

Srivastava et al. (2008, 2009a) conducted source apportionment using CMB and PCA for coarse and fine fractions of PM_{10} and found vehicular exhaust (62%) and crustal re-suspension (35%) to be the dominant contributors to the total ambient concentration in the fine size range of PM while crustal dust (64%) and vehicular pollution (29%) were found to be contributing to the coarse size range using CMB. Analysis of the same data using PCA revealed that crustal re-suspension and vehicular pollution contribute to the coarse range while vehicular pollution (86%) is the major contributor followed by crustal re-suspension (10%) are the key contributors in the case of the fine size range. For the coarse range, the results were found to be more or less similar but in the case of fine range fraction, while PCA attributed 85% to vehicular emissions, CMB attributed only 62% (Srivastava et al., 2008). The results of the analysis in terms of source identification and apportionment are in qualitative agreement with earlier results obtained by Balachandran et al., (2000) and Khillare et al. (2004) with vehicular/industrial sources and crustal re-suspension as discussed earlier.

Since most authors have used different source categories and have sampled different size fractions of PM, it is difficult to inter-compare the results of the various studies. Road dust/soil dust/re-suspended dust has been found to make a substantial contribution to the coarse fraction of PM, particularly in the summer season. While Khillare et al. (2004) and Shridhar et al. (2010) reported similar figures of 22% of variance and 25% of variance respectively, Srivastava et al. (2008) reported 67% of the variance for the coarse fraction of TSPM to be due to re-suspended dust. Only about 10% was found to be contributing to the TSPM fine fraction (Srivastava et al., 2008). Balachandran et al. (2000) reported this factor to be explaining 15.7% of the total variance for PM_{10}. Also, Tiwari et al. (2009) reported two factors associated with re-suspended dust for PM_{10}, explaining 27% and 10% each. Shridhar et al. (2010) also reported re-suspended material explaining 35% of the variance for rural site in Delhi. For $PM_{2.5}$, while Tiwari et al. (2009) reported one factor explaining 6% of the variance, Chowdhury et al. (2007) attributed between 11-42% of the total $PM_{2.5}$ mass across different seasons with the highest figures for summer. The industrial emissions have been estimated to contribute approximately 100-900 tons of $PM_{2.5}$/year (Sahu et al., 2011). Balachandran et al. (2000) attributed 19.4% variance of the PM_{10} mass to industrial emissions while Shridhar et al. (2010) attributed 20% of the variance of the TSP mass to the industrial emissions for the urban site. In some studies, vehicular and industrial emissions have been combined and used as one source category. For example, Balachandran et al. (2000) attributed 53.9% variance of the PM_{10} mass while Khillare et al. (2004) attributed 60% of the variance of the TSP mass to the same source. Biomass combustion is known to be a major source for PM in the city, particularly in winter. Chowdhury et al. (2007) reported that biomass combustion contributes between 7-20% to the total $PM_{2.5}$ mass while Shridhar et al. (2010) reported that 10% of the variance for TSP is due to biomass burning.

CPCB (2010) reported the major sources of PM_{10} as combustion sources, soil dust, re-suspended dust, vehicular emissions, industrial emissions, smelters and secondary aerosols based on factor analysis results. On the other hand, CMB analysis for PM_{10} attributed 5-40% to vehicular emissions, 12-52% to open burning, 7-37% to DG sets, 14-50% to construction, 11-19% to industries and 17-79% to re-suspended dust across the ten sampling sites and different seasons (summer, winter, post-monsoon).

Source Apportionment of Particulate Matter in other Indian Cities

Receptor modelling studies have been conducted in a range of other Indian cities using a range of different methods including CMB and multivariate analysis models. Wind-blown dust and wood/coal combustion were reported as the key sources of particulate matter (TSP) in Mt. Abu in Rajasthan (Negi et al., 1996). Bandu et al. (2000) conducted source apportionment using microscopic methods in Chandigarh and identified soil dust as the key source with others sources including industrial emissions, vehicular traffic and refuse burning. Vehicular traffic was not found to be contributing much to the particulate matter concentration in the city. Kumar et al. (2001) analysed particulate matter concentrations from two different traffic intersections in Mumbai and identified five potential sources including road dust, vehicular emissions, marine aerosols, industries (metal) and coal combustion while Tripathi et al. (2004) identified crustal/ road dust (69.41%), industrial emissions (11.76%) and fuel oil combustion (6.52%) as the key sources. Chelani et al. (2008) analysed samples using inorganic markers from different sites types and identified crustal factor, industrial emissions, vehicular emissions, and marine aerosols as the key sources with the proportions varying at the different sites. Motor vehicles (29%), industrial emissions (23%), two-stroke emissions with fugitive dust (18%), sea salt (9%) and soil (3%) have been identified as the key sources of $PM_{2.5}$ in Navi Mumbai (Maharashtra) using FA-MLR (Kothai et al., 2008).

Sharma and Maloo (2005) reported high concentrations of $PM_{2.5,}$ metals and benzene-soluble organic fraction in the ambient PM_{10} concentration in Kanpur. Gupta et al. (2007) used CMB for source apportionment for SPM and PM_{10} at residential and industrial sites in Kolkata, West Bengal and reported coal combustion and vehicular emissions as the major contributors to PM_{10} at the residential and industrial sites respectively. Other contributors included wood combustion, field burning, solid waste, tyre wear and soil and road dust. In 1995, key sources contributing to atmospheric aerosols in Agra were listed as crustal sources, industrial emissions, wood burning and coal combustion in brick kilns (Kulshrestha et al., 1995) whereas in 2009, Kulshrestha et al. (2009) identified re-suspended dust (due to vehicular activity), solid waste incineration and industrial emissions to be key contributors to the metallic fraction of particulate matter at an urban site in the city of Agra whereas re-suspension, construction activities and industrial emissions were found to be the key sources of PM in the rural location using PCA. Khare and Baruah (2010) analysed sources of $PM_{2.5}$ in Jorhat, Assam using enrichment factor analysis and absolute PCA and reported traffic induced crustal sources (38%), coal combustion (26%), industrial and vehicular emissions (19%), wood burning (9%), and secondary aerosol formation (8%) as the key sources.

Gummeneni et al. (2011) analysed PM_{10} and $PM_{2.5}$ concentrations in a traffic corridor in Hyderabad and identified resuspended dust, vehicular pollution, combustion, industrial emissions and refuse burning as major sources with re-suspended dust being dominant in the case of PM_{10} and vehicular pollution being dominant in the case of $PM_{2.5}$.

Detailed information about receptor model and tracer type and chemical component concentrations from the studies is presented in Table S1.

Central Pollution Control Board (CPCB) Source Apportionment Study

The Central Pollution Control Board (CPCB, 2010) recently released the six-city source apportionment analysis that was initially commissioned as a part of recommendations of the Auto Fuel Policy (2003). A number of research institutions including TERI, NEERI, ARAI, IIT-Kanpur and IIT-Chennai were involved in the analysis, which included pollutant monitoring, preparation of emission inventories and receptor and dispersion modelling. The results of the analysis have been used to prepare city-specific action plans, and may also feed into the next auto policy of India.

Though the methodology used for the study is robust, and can help in conclusive determination of action points, the execution has been poor, and there are many gaps that need to be filled before using the information for actual implementation.

One of the major considerations in conducting any source apportionment study using multivariate methods is to ensure a high ratio between the number of samples collected and the number of variables being analysed, and if that ratio is too low, the results can be misleading. For example, in the case of Chennai, data from 30 samples were analysed for 18 variables. Also, the results obtained from CMB and FA are in many cases not consistent with each other. For example, in Delhi, while the CMB results indicated construction dust to be a major source at several locations, results from factor analysis do not include that source at all. Similarly, the use of tracer markers is ambiguous in several cases including the use of only OC, $SO_4^{2-,}$ EC and TC for identification of auto exhaust or the use of Ni, Na and Si for soil dust as opposed to Al, Ba, Ca, Mg, Mn, Fe, OC, Si and Sr for re-suspended dust. In another case, only the Mg^{2+} ion has been used to identify the soil dust component for one of the sites in Delhi, and only TC and EC are used to identify auto exhaust at another site.

As discussed above, a number of studies have been conducted previously for source apportionment of PM_{10} in New Delhi, Mumbai etc and it is interesting to note that the results of the current analysis are at variance with the earlier reports. While some of the conclusions are similar in both cases (e.g. percentage contribution of road dust to PM_{10} concentrations), there are a number of differences in terms of apportionment of particulate matter to various sources. For example, none of the previous studies have identified domestic LPG use as a source of PM_{10} in New Delhi. Also, very limited analysis was conducted on $PM_{2.5}$ and given its importance in terms of human health impacts, more detailed analysis needs to be conducted.

DISCUSSION AND CONCLUSIONS

There have been many studies conducted in India using receptor modelling methods for source apportionment of particulate matter. India is a very large and diverse country, and unsurprisingly the studies have drawn widely differing conclusions. Even within individual Indian cities, different authors have come to widely varying conclusions over source attribution and apportionment, and this may to some extent be a result of using different sampling locations and seasons. Most studies have identified vehicle emissions and soil/road dust as a major contribution to the fine and coarse fractions respectively, but differentiation of these from industrial emissions and other sources such as construction activity has been poor.

The studies conducted to date do not inspire any degree of confidence that representative knowledge of the source apportionment of any PM size fraction has been gained for any Indian city. The reasons for this view are multiple and include the following:

- The vast majority of studies have used multivariate statistical methods which have yielded factors represented by combinations of elemental and ionic constituents which cannot be unequivocally attributed to any specific source. Faced with factors associating often strange combinations of chemical components, authors feel obliged to attribute a source, but in many cases these are highly unconvincing. The possible reasons are many and include genuine collinearity of sources, or more likely an inadequate number of samples relative to the number of analytes leading to instability in the statistical model. This may be seen in Table S1 which shows that most of the studies used well under 100 samples. It is recommended that future studies using multivariate statistical methods collect at least 100 samples and preferably more.
- The use of unweighted models such as PCA leads to less adequate factor resolution than more recent weighted models such as PMF. The latter is strongly recommended for use in future studies.
- Failure in most cases to distinguish vehicle exhaust from non-exhaust vehicle emissions, particularly resuspension of road dust, and/or inability to differentiate regional crustal sources (e.g. desert dust) from local wind-blown soils and from resuspended road dust. Making a distinction between road dust and local soils can be difficult under any circumstances if the soils are polluted by vehicle emissions or the road dusts contain a significant soil contribution. However, separating these sources, and in particular quantifying the vehicle exhaust contribution alone, and differentiating regional crustal sources from local soils and road dust, is crucial, as the policy response depends heavily upon these insights.
- Most studies pay little attention to secondary pollutants. Sulphate, which in developed countries is almost exclusively secondary, tends to be attributed to local primary sources, and regional transport processes are largely ignored. Similarly, nitrate receives little attention despite its complex atmospheric chemistry and frequent association with regional processes in developed countries (Abdalmogith and Harrison, 2005). Secondary

organic aerosol may be an important contributor to PM mass in India as the conditions exist to facilitate its formation from both anthropogenic and biogenic precursors, but the literature ignores it.

- There is a lack of multi-site studies. Where these exist, they tend to use multiple sites within a city (e.g. CPCB, 2010) rather than using urban/rural contrasts to elucidate the importance of emissions within the city relative to the regional background.
- Emissions inventory data are very scarce. These need to be spatially and chemically disaggregated. Knowledge of city-specific emissions inventories for specific chemical components would give greater confidence in assigning sources to factors identified through multivariate receptor models.
- There has been insufficient use of size fractionation of particulate matter. Most studies have focussed upon TSP or PM_{10}, therefore not benefiting from the additional insights to be gained from separating coarse from fine particles, and in doing so achieving a crude separation of crustal/soil/road dust/construction sources from those associated with high temperature processes (fuel combustion, metallurgical industries, etc) and gas-to-particle conversion to form secondary pollutants.
- There has, to date, been insufficient use of organic molecular markers. While these alone will not answer all source apportionment questions, they are an important tool in receptor modelling and could help to sharpen up both CMB and multivariate model studies.
- Given the rather weak performance of multivariate receptor model studies in India, a greater use of CMB methods is recommended. This will require the use of locally determined source profiles, as well as use of emissions inventories to identify those source types which need to be included. The Central Pollution Control Board study (CPCB, 2010) has generated useful data in regard of local source profiles, but this may need to be complemented by additional measurements.
- Source apportionment based upon emissions inventories and dispersion models is a valuable complement to receptor modelling. Progress in this respect was made by the Central Pollution Control Board study (CPCB, 2010), but it is disappointing that that study failed to develop explanations for the frequently large divergences between modelled and measured PM concentrations. The study also used both multivariate statistical and CMB source apportionment models, but failed to reconcile the sometimes highly divergent conclusions of the two approaches.

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TABLE CAPTIONS

Table 1: Comparison between CMB and multivariate models (based upon Gordon, 1980; Henry et al., 1984; Thurston and Lioy, 1987; Harrison et al., 1997; USEPA, 1997; Viana et al., 2008a; Zeng et al., 2010)

Table 2: Source signatures used for source identification in Indian studies

Table 3: Heavy metal emission sources in the UK (Vincent and Passant, 2006; NAEI, 2011)

Table 1: Comparison between CMB and multivariate models (based upon Gordon, 1980; Henry et al., 1984; Thurston and Lioy, 1987; Harrison et al., 1997; Shrivastava et al., 2007; USEPA, 1997; Viana et al., 2008a; Zeng et al., 2010)

CMB Model	**Multivariate Models**
A key prerequisite is detailed information about the sources/emission inventories	Qualitative information about the potential sources is enough, useful for areas where detailed emission inventories are not available
Only one sample is required	Require large numbers of samples
Does not apportion the secondary aerosols	
Cannot take into account the time variation of the pollutant concentration or source emission	Unable to account for spatial and temporal correlation between emissions (e.g. motor vehicle and road dust) or source identified may contain more than one source
Only non-reactive, stable tracer species can be used	
	Often unable to produce a fine resolution of the sources
Near collinearity among source profiles can result in negative source contributions	Some of the models allow negative contributions to sources which is physically impossible (e.g. PCA)
	Information like met data, particle size etc can be incorporated in the analysis

Table 2: Source signatures used for source identification in Indian studies

Source Signature	Size fraction	Identified Source	City	Reference
Na, Mg, K	SPM	Marine	Mumbai	Kumar et al. (2001)
Na, K	PM_{10} divided into coarse and fine fractions		Mumbai	Kothai et al. (2008)
Na, Cl	PM_{10}		Mumbai	Chelani et al. (2008)
K	SPM	Biomass burning	Delhi	Shridhar et al. (2010)
K, NH_4^+	$PM_{2.5}$		Jorhat	Khare and Baruah (2010)
As, SO_2	SPM	Coal combustion	Mumbai	Kumar et al. (2001)
Co	TSP		Delhi	Srivastava and Jain (2007b)
Picene	$PM_{2.5}$		Delhi, Mumbai, Kolkata	Chowdhury et al. (2007)
Te, S, Mn, Sn, Sb, SO_4^{2-}, Cd	$PM_{2.5}$		Jorhat	Khare and Baruah (2010)
Zn, Pb	TSP	Refuse/ solid waste burning	Bombay (Mumbai), Bangalore, Nagpur, Jaipur	Negi et al. (1987)
TC, OC, NO_3^-	PM_{10}		Kolkata	Karar and Gupta (2007)
Cr, Ni	PM_{10} divided into coarse and fine fractions		Navi Mumbai	Kothai et al. (2008)
Cu, Cr, Ni	$PM_{2.5}$ and PM_{10}		Agra	Kulshrestha et al. (2009)
Zn, Pb	PM_{10}		Delhi	Chelani et al. (2010)
Coarse mode of Pb, Cr and fine mode of Pb, Cr, Ni	PM_{10} divided into coarse and fine fractions	Vehicular/industrial	Delhi	Balachandran et al. (2000)
Cd, Cr, Ni, Pb	SPM		Delhi	Khillare et al. (2004)
Pb, Co, Sb	PM_{10} divided into coarse and fine fractions		Navi Mumbai	Kothai et al. (2008)
Co, Ni, Cu, Zn, Cd, Te	$PM_{2.5}$		Jorhat	Khare and Baruah (2010)
Zn, Cu, Br,V, Mn	TSP	Industrial	Bombay (Mumbai), Bangalore, Nagpur, Jaipur	Negi et al. (1987)
Coarse and fine modes of Ni, Cd	PM_{10} divided into coarse and fine fractions		Delhi	Balachandran et al. (2000)
Cu, Mn, Ni	SPM		Mumbai	Kumar et al. (2001)
Fe, Se, Mo, Sb with low loading of Cr, V, Mn, Cu	PM_{10}		Tirupati	Mouli et al. (2006)
Mn, Cr	$PM_{2.5}$ and PM_{10}		Agra	Kulshrestha et al. (2009)
Ni, Cd, Ba, Na	PM_{10}		Delhi	Chelani et al. (2010)
Ni, Cr, Mn, Cu, Zn, Pb	TSP		Delhi	Shridhar et al. (2010)
Co, Cr, Zn, Cu, Ni	TSP		Kolkata	Kar et al. (2010)
Pb	TSP	Vehicular	Bombay (Mumbai), Bangalore, Nagpur, Jaipur	Negi et al. (1987)
Pb, Cr, NO_2	SPM		Mumbai	Kumar et al. (2001)
Cu, Pb, Cr	TSP		Delhi	Srivastava and Jain (2007b)
Cr, Pb, Ni, Mn, Fe	PM_{10}		Kolkata	Karar and Gupta (2007)
Zn, S, BC	PM_{10} divided into coarse and fine fractions		Navi Mumbai	Kothai et al. (2008)
Cu, Cr, Zn, Ni, Ca, Zn, Co	PM_{10} divided into coarse and fine fractions		Delhi	Srivastava et al. (2008)
Cu, Zn, Pb	PM_1		Kanpur	Chakrobarty and Gupta (2010)
Pb, Ni, Mn, Zn, Cu	$PM_{2.5}$ and PM_{10}		Agra	Kulshrestha et al. (2009)
Cu, Ni, Co, Cr, Ca	TSP		Delhi	Srivastava et al. (2009a)
Zn	SPM		Mithapur	Basha et al. (2010)
EC, OC, Zn	PM_{10}		Delhi	Chelani et al. (2010)
Al, Si, Ca, Ti	TSP		Bombay (Mumbai), Bangalore, Nagpur, Jaipur	Negi et al. (1987)
Fine and coarse mode of Fe	PM_{10} divided into coarse and fine		Delhi	Balachandran et al. (2000)

	fractions	Crustal dust/ road dust/ re-suspended dust		
Fe, Al, Ca	SPM		Mumbai	Kumar et al. (2001)
High loading of Fe	SPM		Delhi	Khillare et al. (2004)
Li, V, Cr, Mn, Ni, Co, Cu, Pb	PM$_{10}$		Tirupati	Mouli et al. (2006)
Cl$^-$, SO$_4^{2-}$	PM$_{10}$		Kolkata	Karar and Gupta (2007)
TC, OC, Fe	TSP (road dust)		Kolkata	Gupta et al. (2007)
OC, TC, Cl$^-$, SO$_4^{2-}$	TSP (soil dust)		Kolkata	Gupta et al. (2007)
Ni, Pb, Mn, Fe, Cu, Cd, Cr, Co	TSP		Delhi	Srivastava and Jain (2007b)
Fe, Cd, Zn, Ca, Cr, Ni, Mg, Ca	PM$_{10}$ divided into coarse and fine fractions		Delhi	Srivastava et al. (2008)
Fe, Sc, Si, Ti, Ca	PM$_{10}$ divided into coarse and fine fractions		Navi Mumbai	Kothai et al. (2008)
Mn, Mg, Fe, Al, V, Co	PM$_{10}$		Mumbai	Chelani et al. (2008)
Cr, Zn, Cd, Mg, Ca	TSP		Delhi	Srivastava et al. (2009a)
Ca, Mg, Fe, Pb with moderate loadings of Zn, Cr, V	PM$_1$		Kanpur	Chakrobarty and Gupta (2010)
Fe, Ni, Zn, Pb	PM$_{2.5}$ and PM$_{10}$		Agra	Kulshrestha et al. (2009)
Al, Si, Ca, Ti	PM$_{2.5}$		Jorhat	Khare and Baruah (2010)
Ca, Mg, Fe, Al, Mn	SPM		Delhi	Shridhar et al. (2010)
Pb, Cr, Co	SPM		Mithapur	Basha et al. (2010)
Fe, Cr, Mn, K$^+$	PM$_{10}$		Delhi	Chelani et al. (2010)
Ca, Co	TSP	Construction activities	Delhi	Srivastava and Jain (2007b)
Fe, Pb, Ni	PM$_{2.5}$ and PM$_{10}$		Agra	Kulshrestha et al. (2009)
Cd, V	SPM	Refuse oil burning	Delhi	Shridhar et al. (2010)

Table 3: Heavy metal emission sources in the UK (Vincent and Passant, 2006; NAEI, 2011)

Source type	**Tracer elements**
Coal combustion (domestic/industrial) +electric arc furnaces + coke ovens	As, Cd, Cr, Cu, Pb, Mn, Mg, Ni, K, Se, Zn
Industrial emissions	Cd (copper alloy manufacture, Al, Zn production, galvanizing, lead acid battery industry), Cr (Production of Cr-based chemicals), Cu, Pb (industrial metal, lead-acid battery), Mn, Ni (Al production, refineries, copper alloy manufacture)
Fuel oil combustion	As, Cd, Cu, Mn, Ni, V
Iron and steel production	Cr, Pb, Mn, K, V, Zn
Road transport	Cu and Zn (tyre and brake wear), V (diesel)
Foundries	As, Cd, Pb, Ni
Waste burning/ incineration	Cd, Cu, Pb, Zn