PM$_{2.5}$ Source Apportionment of Major Cities in Asia

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Contents

Introduction

This report was designed to report the current best understanding of the sources contributing to $PM_{2.5}$ in major cities around the world in recent years (sampling in 2015 or later). Prior work (Hopke et al., 2020) provides an inventory of all source apportionment studies published between 2015 to August 2019, but most of the actual ambient aerosol sampling occurred prior to 2015. In the period of 2014 to 2019, there have been significant reductions in emissions in many countries as a result of the implementation of emissions control policies. Of particular note has been actions taken in China as a result of the 2013 National Air Quality Action Plan and the subsequent Coal-to-Gas program to convert northern China from coal combustion. Programs in other countries such as the United States continued the implementation of existing programs such as the transition from Tier 2 light duty vehicle standards to Tier 3 standards starting on January 1, 2017. Thus, apportionments made on PM samples collected earlier in the period are unlikely to fully reflect the relative contributions of those sources to the current $PM_{2.5}$ concentrations. Thus, this report has focused on published studies that relied on the data from the analysis of the most recently collected samples. Beginning in 2020, the onset of the COVID-19 pandemic resulted in many changes in the air quality system with reduced emissions during periods of enforced reduced activities (Amouei Torkmahalleh et al., 2021). However, there have been very different approaches to the continuing infection threats that made it difficult to collect data and its applicability to the likely post-pandemic system is unclear. Thus, this report focuses on the pre-pandemic results.

The objective was to examine the source apportionment in 31 large cities from around the world (Table 1). However, searches on Google, Google Scholar, Scopus, and the Web of Science for $PM_{2.5}$ and "source apportionment" only identified 20 of these cities with useful reports of source apportionments. Thus, the following sections provide analyses of each city based on one or a few papers that provide the best currently available source apportionments and include assessments of the likely accuracy of what is reported.

Table 1. Cities in which recent source apportionment results were assessed.

Bangkok (no reports)	Islamabad (no reports)	New Delhi
Beijing	Jakarta	Phnom Penh (no reports)
Berlin (no reports)	Karachi	Seoul
Chengdu	Kathmandu (no reports)	Shanghai
Colombo (no reports)	Kuala Lumpur	Shenzhen
Dhaka	London (no reports)	Singapore
Guangzhou	Los Angeles (no reports)	Tokyo (no reports)
Hangzhou (no reports)	Manila	Ulaanbaatar
Hanoi	Mumbai	Vientiane (no reports)
Hong Kong	Nanjing	Wuhan
		Xi'an

China

Beijing

There have been major changes in the sources contributing to particulate pollution in Beijing since 2013 when the National Air Quality Action Plan was initiated. As a point of comparison, Li et al. (2019a) analyzed particulate pollution data from June 2012 to April 2013 including the very high period in January 2013. Their identified sources were: Traffic, Biomass Burning, Nitrate/Sulfate, Incineration, Sulfate, Coal Combustion/Ammonium Chloride, Residential Coal/Biofuel Combustion, and Road Dust/Soil with mass contributions (fractions) to $PM_{2.5}$ of 12.35 (10.4%), 8.70 (8.9%), 24.51 (22.4%), 5.64 (7.2%), 25.14 (24.5%), 7.10 (6.2%), 14.18 (15.4%), and 5.33 $\mu g/m^3$ (5.0%), respectively. Du et al. (2022) analyzed $PM_{2.5}$

compositional data from samples collected at a site in the Chaoyang district from 2013 to 2018. However, few details of the analyses are presented since they do not provide the time series of resolved source contributions. They used a very limited set of 10 chemical species and resolved only 6 source types: Road dust, Vehicle exhaust, Coal combustion, Biomass burning, Secondary nitrate, and Secondary sulfate. The average contributions of these sources were 1.77 (3.12%), 11.04 (19.47%), 6.26 (11.04%), 3.10 (5.47%), 16.01 (28.23%), and 17.05 µg/m^3 (30.07%), respectively. The data do not include nitrate and have substantial issues with their profiles such as having both Cl^- and K^+ in the same "biomass" profile. During this period, there would have been significant amounts of coal combustion that includes Cl^- as a major tracer (Yu et al., 2013). There have been other short-term source apportionment studies in Beijing, but there has not been reports of a study over the 2013 to 2019 time period documenting the changes in source profiles and/or emissions resulting from major changes such as the implementation of improved controls on large scale emitting facilities such as coal-fired power plants and industrial factories. Beginning in 2016-17, there was an intensive effort to convert the heating facilities in Beijing from coal to natural gas combustion.

A more recent analysis has been reported by Park et al. (2022) based on 2019 data collected at the Chinese Research Academy of Environmental Research (40.042°N, 116.413°E). This work collected multiple filters and performed a more complete analysis of the collected filter samples providing 17 trace elements, 6 ions, and organic and elemental carbon (OC/EC). They applied dispersion-normalized Positive Matrix Factorization (DN_PMF) (Dai et al., 2020a; Chen et al., 2022) to the data. The concept of DN-PMF is that the PM concentrations are driven by both emission rates and atmospheric dispersion. Thus, during periods of higher winds and/or higher boundary layer heights, the same rate of emission will produce a lower ambient concentration and vice versa. Thus, by normalizing the data with the ventilation coefficient (VC = wind speed x boundary layer height) to the average VC over the whole sampling period, the effects of the variations in dispersion will be substantially reduced. The improvement was particularly clear in the analysis of hourly data (Dai et al., 2020a), but it still made notable improvements for 24 hour integrated data (Chen et al., 2022).

The $PM_{2.5}$ mass concentrations ranged from 5.1 to 165.1 µg/m^3 and the average mass concentration of $PM_{2.5}$ was 41.9 ± 29.7 µg/m^3 (Table 1). The average mass concentrations of $PM_{2.5}$ were the highest in winter (55.7 ± 37.1 µg/m^3) followed by spring (47.4 ± 38.2 µg/m^3), autumn (45.8 ± 27.9 µg/m^3), and summer (31.1 ± 15.6 µg/m^3). The average annual concentrations of NO_3^-, SO_4^{2-}, and NH_4^+ were 11.0 ± 13.0 µg/m^3, 5.7 ± 4.4 µg/m^3, and 4.4 ± 4.3 µg/m^3, respectively, and these major ionic species accounted for 50.5% of the total mean $PM_{2.5}$ mass concentration. The source apportionment results were based on 224 samples collected throughout the year and are presented in Table 2.

There are notable differences between the 2012-13 results (Li et al., 2019a) and the 2019 values (Park et al., 2022). In January 2013, sulfate was higher than nitrate that is quite unusual. Given that NO_2 reacts 10 times faster with hydroxyl radical than SO_2, most western locations see high particulate nitrate in the winter and high secondary sulfate in the summer. In 2019, there is still relatively high winter sulfate, but nitrate has become the dominant secondary species. Li et al. (2019a) attributed a substantial contribution to both sulfate and organic carbon from residential solid fuel (mostly coal) combustion. Residential coal burning in typical household stoves gives rise to the emission of primary sulfate (Dai et al., 2018) and primary oxidized organic carbon including humic-like substances (HULIS) (Li et al., 2019b). The difference between January 2013 and winter 2019 is likely due to the effects of the coal-to-gas program that started in Beijing in 2016-17. District heating systems were converted to natural gas as were a number of the home in the peri-urban areas of Beijing. Coal combustion has also been substantially reduced in the 2019 results and a separate residential heating/cooking source was not resolved. Thus, for the immediate area around Beijing, the combination of improved controls on coal-fired power plants and industrial emissions that occurred in 2014 to 2015 along with the coal-to-gas conversions on 2016-17 have substantially improved Beijing particulate air quality especially in winter. However, the remaining concentrations are still far from the new 2021 WHO air quality guidelines (WHO, 2021) to more fully protect public health.

Table 2. Contribution of the sources resolved by DN-PMF in Beijing in 2019 (Park et al., 2022).

Source	Concentration (µg/m^3)	Percent Contribution
Secondary Nitrate	13.063	31.7%
Secondary Sulfate	7.225	17.6%
Traffic	4.717	11.5%
Oil Combustion	2.310	5.6%
Coal Combustion	2.642	6.4%
Soil	2.807	6.8%
Incinerator + Industry	4.208	10.2%
Biomass Burning	3.599	8.7%
Aged Sea Salt	0.593	1.4%

Chengdu

Kong et al. (2020) analyzed hourly $PM_{2.5}$ mass concentrations and chemical species measure at the Chengdu Academy of Environmental Sciences at 25 m above the ground. The area includes roads, commercial and residential areas with no buildings taller than 200 m. The annual average $PM_{2.5}$ concentration in Chengdu was 67.44 ± 48.78 mg/m^3. The average $PM_{2.5}$ mass concentrations in spring, summer, autumn, and winter were 58.03 ± 27.28, 41.45 ± 16.87, 55.48 ± 32.34, and 115.41 ± 65.28 µg/m^3, respectively. There were episodes of very high concentration including December 30, 2015 when the $PM_{2.5}$ concentration reached 311.7 µg/m^3. They resolved 6 sources that were assigned as motor vehicle emissions (8%), biomass burning (11.7%), industrial sources (3.1%), secondary aerosols (35.9%, coal combustion (27.3%), and dust (13.9%). Given one year of data, it would have been expected that there would have been sufficient differences in sulfate and nitrate concentrations that they could have been resolved rather than being assigned to the same factor. The covariance between sulfate and nitrate should have been reduced by the low particulate nitrate in the summer and lower sulfate production during the winter. This issue suggests that they did not adequately explore the range of factor numbers to test whether these two major secondary inorganic species could have been separated. Coal combustion and biomass burning clearly increase during the period for which space heating was needed with its highest contribution in winter. Vehicular contributions had an unusual seasonal pattern with the highest concentration in summer, followed by spring, autumn, and winter. Given that in winter, dispersion conditions are poorest and traffic volumes generally do not vary widely with season, this pattern raises doubts about the results.

Recently, Xue et al. (2022) reported the results of a study based on 80 quartz filter samples collected in 2018. Seasonal samples were collected in April, July and August, October and November, and January through February. Samples were not collected on rainy or snowy days. They analyzed the samples for the typical 7 ions, 18 elements, OC/EC. However, they also extracted them and analyzed for PAHs, hopanes and steranes using gas chromatography-mass spectrometry. They applied PMF to only the standard species with PAHs and again after including the additional organic marker compounds providing 57 species in the 80 samples. Six factors were identified by incorporating conventional components and PAHs, including secondary source (SS) (28%), fugitive dust (FD) (15%), construction dust (CD) (4%), coal combustion (CC) (13%), gasoline vehicles (GV, 12%), and diesel vehicles (DV, 10%). Eight factors were identified by incorporating conventional components, PAHs, hopanes, and n-alkanes, including SS (26%), FD (17%), CD (3%), GV (14%), DV (8%), immature coal combustion (ICC, 5%), mature coal combustion (MCC, 10%), and biogenic source (BS, 1%).

They were unable to separate sulfate and nitrate even with seasonal samples that by 2018 would have been expected to provide sufficient differences between winter nitrate and summer sulfate to permit separation into two factors. Given important differences in sources and the replacement of nitrate in the $PM_{2.5}$ as sulfate was reduced thereby freeing ammonia to drive the ammonia-nitric acid equilibrium to the particulate phase, it is disappointing that these two major source types could not be resolved. Recent work on the application of dispersion normalization to daily sample data (Chen et al., 2022) suggest that this additional step prior to the PMF analyses might improve the resolution of these seasonally different source types.

With the inclusion of the additional molecular marker species, they do separate two types of coal burning: "mature" and "immature". The "mature" coal combustion included OC, EC, $SO_4^=$, Si, Ca, Flt, Pyr, BaA, BbF, 17α(H),21β(H)-30-norhopane, 17α(H),21β(H)-hopane, C24, and C27. Anthracite is "mature" coal. Alternatively, the "immature" coal combustion is characterized by OC, EC, $SO_4^=$, Si, Ca, Flt, Pyr, BaA, BbF, 17ß(H),21ß(H)-hopane, C16, C18, and C25. The other types of coal (bituminous, subbituminous, and lignite) represent "immature" coal. A specific compound, 17α(H), 21β(H)-30-norhopane, is used to identify the maturity of the coal being combusted (Bi et al. 2008; Oros and Simoneit 2000; Tian et al. 2021). 17ß(H), 21ß(H)-hopane, C16, and C18 are regarded as markers of immature coal combustion source (Bi et al. 2008; Choi et al. 2015; Lin et al. 2010; Wang et al. 2009, 2015). They have separated the coal types but not the actual combustion systems (coal-fired power plants, coal-fired industrial boilers, and residential coal combustion). Residential coal combustion is significantly different from the other two source types since lump coal or briquettes are used rather than pulverized coal. The large pieces of fuel result in greater difficulty in high temperature combustion leading to significant emissions of primary sulfate (Dai et al., 2019) and oxidized primary organic compounds including HULIS (Li et al., 2019b).

The availability of hopane concentrations permits the separation of diesel from gasoline vehicles given much higher emissions of hopanes and steranes from diesel vehicles (Schauer et al., 1996). The longer chain alkanes values permit the identification of primary biological materials, typically leaf fragments, given that the wax on the leaves has a distinctive odd-even carbon chain length pattern (Schauer et al., 1996).

Hong Kong

There is a recent report of source apportionments for Hong Kong (Chow et al., 2022). Chow et al. (2022) reported the apportionment of $PM_{2.5}$ collected at 6 sites across Hong Kong. Samples were collected every 6 days from January to December 2015 with a mid-volume sampler collecting on a 47 mm Teflon filter and a 47 mm quartz fiber filter. A high-volume sampler was used to collect $PM_{2.5}$ on 8′ × 10′ quartz fiber filters. A total of 317 sets of filter samples were collected in their campaign. Elements on the Teflon filters were determined by XRF. OC/EC and water-soluble ions were determined from the 47 mm quartz filter. Portions of the high-volume sampler quartz filters were leached for levoglucosan, mannosan, erythritol, galactosan, arabitol, sorbitol, mannitol, and glucose. Another portion was extracted and analyzed for non-polar organic compounds.

Nine sources were resolved using PMF that were named: secondary sulfate, secondary nitrate, industrial/coal combustion, biomass burning, primary biological particles, vehicle, residual oil, dust, and aged sea salt. The results of the apportionment are provided in Table 3. The organic tracers permitted additional sources to be resolved relative to what would be possible without them and they provide the results for both with and without the molecular markers. They provide results with respect to the influence of local versus distant sources and report the apportionment of OC and EC as well as $PM_{2.5}$. They ran the PMF diagnostics, but do not report the detailed DISP results (Paatero et al., 2014) for each species in each profile so that we can see the rotational ambiguity present in the profile values.

Table 3. Source apportionments at the 6 monitoring sites in Hong Kong based on samples collected in 2015 (Chow et al., 2022a).

Source	Roadside	Urban	Urban	Urban	Suburban/rural	Suburban/rural
F1 Secondary sulfate	8.84 (31%)	10.16 (40%)	8.61 (35%)	10.42 (46%)	10.03 (46%)	11.03 (49%)

F2 Secondary nitrate	3.58 (13%)	3.68 (15%)	2.33 (10%)	2.35 (10%)	1.32 (6%)	1.33 (6%)
F3 Industrial/coal combustion	3.34 (12%)	5.08 (20%)	3.39 (14%)	4.05 (18%)	3.60 (16%)	3.51 (16%)
F4 Biomass burning	1.44 (5%)	0.70 (3%)	3.15 (13%)	0.64 (3%)	1.67 (8%)	0.49 (2%)
F5 Primary biogenic particles	0.65 (2%)	0.06 (0%)	0.71 (3%)	0.11 (0%)	0.27 (1%)	0.08 (0%)
F6 Vehicular emissions	6.88 (24%)	1.75 (7%)	2.14 (9%)	1.46 (6%)	0.43 (2%)	0.47 (2%)
F7 Residual oil	0.63 (2%)	0.54 (2%)	0.89 (4%)	0.42 (2%)	0.53 (2%)	0.95 (4%)
F8 Dust	1.10 (4%)	1.40 (6%)	1.10 (5%)	1.19 (5%)	1.28 (6%)	1.76 (8%)
F9 Aged sea salt	2.00 (7%)	1.80 (7%)	2.09 (9%)	1.92 (9%)	2.69 (12%)	2.71 (12%)
PMF-apportioned sum	28.5	25.2	24.4	22.6	21.8	22.3
Measured $PM_{2.5}$	29.8	25	23.8	22.5	21.1	22.8

Wuhan

Zhang et al. (2022a) provides a more credible resolution of the $PM_{2.5}$ sources in Wuhan. They collected 123 $PM_{2.5}$ filter samples in Wuhan from December 2014 to November 2015. Analyses were performed to provide concentrations of water- soluble inorganic ions (WSIIs), elemental carbon (EC), organic carbon (OC) and inorganic elements. The annual average $PM_{2.5}$ concentration was 80.5 ± 38.2 µg/m^3 also with higher concentrations in the winter and lowest in the summer. They also used a seasonal sampling strategy with sampling periods included December 8, 2014 to January 12, 2015 (winter), April 24 to May 17, 2015 (spring), August 1 to August 27, 2015 (summer) and October 26 to November 12, 2015 (autumn) with approximately equal numbers of samples collected in each season. The seasonal concentration of $PM_{2.5}$ ranked in the order of winter (109.7 ± 49.1 µg/m^3) > spring (82.8 ± 22.1 µg/m^3) > autumn (82.0 ± 24.7 µg/m^3) > summer (48.6 ± 19.5 µg/m^3). Samples were collected on quartz filters using a medium volume sampler (100 L/min) that were cut into pieces to provide the material for leaching, extraction, or direct measurement of OC/EC. They used a DRI2001 analyzer that produces carbon fraction data (OC1 – OC4, OP, EC1-EC3) that have proven useful in separating diesel from gasoline emissions (Kim et al., 2004), but they did not employ them in their analysis. They report that their elemental analyses were done using XRF on a Teflon filter. However, they do not mention collecting samples on Teflon filters.

PMF resolved five sources including secondary inorganic aerosols (SIA), coal combustion, industry, vehicle emission, fugitive dust. SIA, coal combustion, as well as industry were the dominant contributors to $PM_{2.5}$ pollution, accounting for 34.7%, 20.5%, 19.6%, respectively. With seasonal sampling, it should have been possible to resolve nitrate from sulfate. Employing the full suite of OC/EC data would be expected to resolve diesel from spark-ignition vehicles and possibly other sources so it appears they have not made full use of the available data to apportion the data. They also have performed a cursory exploration of the separation of local versus transported sources that they could have done using local wind data to do Conditional Bivariate Probability Function (CBPF) analysis for local sources and Potential Source Contribution Function (PSCF) or Concentration Weighted Trajectory (CWT) analyses to explore the origins of long-range transported PM (Hopke, 2016)

Zhang et al. (2022b) used a combination of principal components analysis (PCA) and a random forest model to quantify $PM_{2.5}$ sources in Wuhan based on online monitoring from December 2019 to November 2020. $PM_{2.5}$ was the highest in winter (61.33 ± 35.32 µg/m^3) and the lowest in summer (17. 87 ± 10. 06 µg/m^3). Only 5 sources were resolved: coal burning and secondary sources (46%, 39%, 41%, and 52% for spring, summer, autumn, and winter, respectively), vehicular emissions (22%, 28%, 27%, and 21%, respectively), industrial emission sources (14%, 18%, 17%, and 13%, respectively), dust sources (10%, 8%, 6%, and 6%, respectively), and biomass burning sources (8%, 7%, 9%, and 8%, respectively). However, these results are unlikely to be reliable. With hourly data collected over a full year, nitrate and sulfate should be separable separate from coal combustion. PCA and other eigenvector-based methods have been shown by Lawson and Hanson (1974) and Malinowski (2002) to be unweighted least-square fits to the data. Such fits are going to create problems with heteroskedastic data such as is commonly encountered in environmental measurements. It also produces unrealistic orthogonal profiles. As demonstrated by Paatero and Hopke (2003), an eigenvector-based analysis enhances the effect of noisy data and diminishes the impact of higher quality data. There are readily available properly weighted least-squares fitting approaches that will improve the resolution by data point weighting and permit non-orthogonal results. Thus, further analyses of these data are warranted.

Xi'an

Dai et al. (2018) collected $PM_{2.5}$ filter samples at 6 sites in and around Xi'an from December 2014 to November 2015 and analyzed them for water-soluble ions, elements, and OC/EC (Dai et al., 2018). An initial source apportionment was reported by Dai et al. (2019) where they employed the capabilities of EPA PMF V5 to impose constraints on the profiles. In this study, they had measured the composition of the emissions from a typical coal stove burning lump coal or briquettes and constrained the profile initially resolved by PMF to match the measured source composition. However, this paper only reported results related to residential coal combustion (RCC). They found that primary OC from RCC accounted for 41.6% and 31.3% of ambient OC in $PM_{2.5}$ in the urban areas of Xi'an during heating season and non-heating season, respectively. Primary sulfate from RCC, on average contributing 38.9% and 49.2% of ambient sulfate mass in $PM_{2.5}$ at urban sites and rural sites, respectively.

Dai et al. (2020b) then performed a second set of PMF analyses on the same data for 5 of the 6 sites using the availability of multiple site data to set constraints on the source contribution values for the large metallurgical manufacturing facility in Xi'an. With multiple sampling sites, there will be some days when the wind direction is such that one site will intersect the plume while others will not. Constraining the contributions by pulling them toward 0 reduces the rotational ambiguities and should provide a more accurate apportionment of the contributions from that emission source. Three of the sites were affected by the metallurgical facility and they were analyzed together to provide the apportionment. Apportionments were done separately for the other two urban sites.

Seven sources were identified for the multisite PMF analysis; coal combustion, road traffic, soil, biomass burning, sulfate, nitrate, and metallurgical industry. In this case, sulfate and nitrate were separated. The results of the analyses are presented in Table 4.

Table 4. Fractional contributions of the sources resolved in Xi'an using constrained PMF (Dai et al., 2020b).

Site	Fractional Contributions (%)							$PM_{2.5}$ ($\mu g/m^3$)
	Traffic	Biomass Burning	Sulfate	Nitrate	Soil	Coal Combustion	Metallurgical Industry	
Urban	16.3	6.2	18.5	31.4	5.6	17.7	4.3	115.4
Industrial	11.0	5.8	23.0	28.7	5.7	17.3	8.5	117.8
Rural	11.0	7.2	26.3	25.8	4.8	23.1	1.7	113.3
CA[1]	9.9	19.9	15.0	31.0	6.4	17.8	-	110.8
SS[1]	14.2	14.1	11.1	45.0	3.9	11.8	-	106.0

[1] Sites at which the metallurgical industry was not resolved.

A source apportionment study was conducted at 4 sites in the Xixian New District (XXND) that is adjacent to Xi'an (Wang et al., 2021). Daily sampling was done for one month in each of four seasons from 2017 to 2018 resulting in a total of 508 Teflon and 508 quartz filters for chemical characterization. The chemical data for the filters is based on seasons defined as: December 18, 2017–January 21, 2018 as winter, April 27, 2018–May 25, 2018 as spring, July 25, 2018–August 22, 2018 as summer and October 31, 2018–November 29, 2018 as autumn. Water soluble ions, elements, and OC/EC were measured. The data were analyzed using the (CAS Hybrid Environmental Receptor Model, CAS-HERM) developed by the Institute of Earth Environment, Chinese Academy of Sciences, and based on positive matrix factorization (PMF) and chemical mass balance (CMB) receptor models, was used to identify likely pollution sources for the $PM_{2.5}$ (Chen and Cao, 2018). This model is based on multiple misconceptions about the nature of "sources" and the underlying mathematical constructs that go into PMF and chemical mass balance (CMB) analyses. The approach produces results that are overfitted as can be seen by the extremely high r^2 values between the reproduced mass and the measured $PM_{2.5}$ values that are all greater than 0.96. Given the natural variability of the profiles and the inherent uncertainties in the measurements, such high correlations are improbable.

They resolved 7 sources that included industrial processes, motor vehicles, biomass burning, dust-related, secondary sulfate and secondary organic carbon, secondary nitrate and secondary organic carbon, and coal combustion. It is highly unlikely that the OC associated with the sulfate and nitrate factors is only secondary given the emission of semivolatile oxidized primary organic carbon by residential coal combustion (Li et al., 2019b) that will partition onto any particle including primary sulfate (Dai et al., 2019), secondary sulfate, and secondary nitrate particles. In addition, secondary organic aerosol varies widely in composition over the year depending on emissions and oxidation conditions such that the organic mass to organic carbon ratio can vary from 1.6 up to 2.1 (Turpin and Lim, 2001). They found only weak biomass burning contributions with ~10% annually at 2 sites and at the other 2 sites, biomass burning only accounted for ~5%. However, they still should have been reported if they were resolvable.

They then "refine" their results using CMB analyses on the source subsets resolved by the PMF analysis. There is no indication of the origins or reliability of the source profiles used in their analyses and the potential is high for misspecification of the source profiles even if they provide a good fit to the data as shown by Subramanian et al. (2006).

Shanghai

There are a number of partial apportionment studies in Shanghai such as one that focuses on particle bound polycyclic aromatic hydrocarbons (PAHs) or one apportioning PM based only on measured elements that cannot provide accurate results. The only apportionment based on a year of data (2015–2016) is reported by Feng et al. (2022). $PM_{2.5}$ samples were collected every 5 days using high volume samplers with quartz filters on the roof of a 20 m high building from December 2015 to November 2016. The filters were analyzed for organic carbon (OC), elemental carbon (EC), water-soluble ions, and elements. They also measured 16 PAHs: naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorine (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fluo), pyrene (Pyr), ben[a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd] pyrene (IcdP), dibenz[a,h]anthracene (DahA) and benzo[ghi]perylene (BghiP). They resolved 6 factors without the PAHs included and 7 factors when the data included the PAHs. However, there is a serious problem with the results presented in the paper. The DISP intervals they present do not properly overlap with the base run results. In the DISP analysis, the base run profile values are systematically pulled up and down from the base case. The maximum DISP value has to be greater than or equal to the base run value while the minimum DISP value has to be less than the base run value. Thus, either they have plotted the results incorrectly or the values they present are not meaningful and thus, this work will not be further discussed.

There is no other recent complete apportionment based on a sufficiently long data set to be able to report seasonal or longer-term trends. Li et al. (2020a) used a data set based on samples collected between 9 November to 3 December 2018. They measured hourly $PM_{2.5}$ and its chemical components, including water-soluble inorganic ions, carbonaceous material, and trace elements, and organic molecular markers at every odd hour over a 3-week field campaign in urban Shanghai. They identified 11 factors: secondary nitrate (30.4%), secondary sulfate (15.3%), vehicle exhaust (12.6%), industrial emissions/tire wear (3.8%), industrial emission 2 (2.0%), residual oil combustion (2.0%), dust (4.2%), coal combustion (5.3%), biomass burning (4.8%), cooking (2.8%), and secondary organic aerosol (SOA) (16.8%). They also analyzed the data without the molecular markers and then could not resolve the biomass burning, cooking emissions, and SOA factors. The results here are useful, but without seeing seasonal patterns such as spring dust storms, spring agricultural activity that could produce dust or biomass burning if they burn the fields to prepare them for planting, this paper does not provide a full picture of the sources sufficient to develop the best possible control strategy. Further work is clearly needed.

Nanjing

There have been many short-term studies in Nanjing that are detailed in Hopke et al. (2020). Recent studies have focused on apportionment of light absorbing organic carbon, black carbon, water soluble inorganic ionic species, and heavy metals. There is one paper based on one year (2017) of data (Yu et al., 2020). Hourly concentrations of water-soluble inorganic ions ($NH_4^+, SO_4^=, NO_3^-, Cl^-$, Ca^{2+}, Mg^{2+}, K^+, and Na^+) and carbonaceous components (OC, EC) were measured with a MARGA, ADI 2080 and a Sunset Laboratory RT-4, respectively, from January 1, 2017, 0:00 at midnight (local time) to December 31, 2017 23:00. Simultaneous measurements of $PM_{2.5}$ mass, eighteen elements (K, Fe, Zn, Ca, Si, Mn, Pb, Cu, Ti, As, V, Ba, Cr, Se, Ag, Cd, Ni, and Hg), gaseous pollutants (SO_2, NO_2, O_3, and CO), and meteorological variables (ambient temperature, relative humidity, wind direction and speed) were made at the Nanjing Environmental Protection Building (NEPB) site. A 7-factor solution was resolved using EPA PMF V5. The identified sources were: secondary nitrate (37.4%), secondary sulfate (30.8%), on-road traffic (15,1%), coal combustion (7.48%), crustal dust (3.47%), residual oil combustion (2.76%), and metal smelting (2.94%). The way the results are presented makes it difficult to assess the quality of the results. They present the source profiles using a linear scale when species range from major species like sulfate and nitrate to trace elements such as As so it is difficult to fully see the source profiles. They did not provide the explained variation values nor the DISP results for each species in each profile and these are critical to fully assess the quality of their attribution of the factors to source types. The diagnostics presented in Table S3 of their supplemental material file suggests that their solutions were robust. Although they examined the directionality of local sources with bivariate polar plots and conditional probability function analysis, there was no trajectory ensemble work to assess the probable origins of distance sources of factors like secondary sulfate. They extensively examine the temporal variations of the measured species but not the resolved source contributions. Thus, they have not fully exploited the information content in their results.

Xie et al. (2022) analyze data from $PM_{2.1}$ and $PM_{2.1\text{-}10}$ data based on samples collected in northeastern Nanjing, a residential and academic institutional area. They used nine-stage Andersen cascade impactors with one impactor loaded with quartz filters while the other's stages were loaded with PTFE filters. Seventy two-hour samples were collected consecutively in a typical month of each season from 2016 to 2017. However, the total number of samples collected and analyzed is not provided. The Teflon filters were weighed for mass and then cut into pieces to support multiple analyses. One-quarter was water leached for analysis of inorganic ions while another quarter was acid digested and analyzed for elements by ICP-MS. A punch from the quartz filter was analyzed for OC and EC using the IMPROVE-A protocol. The resulting data were analyzed using EPA PMF V5 for the aggregated data from the stages up to 2.1 μm and the data aggregated from the larger size particle stages.

They present their source profiles in an unusual way using linear scales and as percent rather than as μg of species per μg of PM. Thus, trace elements are extremely hard to discern. They ran the EPA PMF diagnostics, but they do not present the DISP intervals so the significance of specific species in each profile can be assessed. The diagnostic results summarized in their Table S5 suggest significant uncertainties in their results with poor correspondence between the bootstrap runs and the base run. They identified a mixed secondary inorganic aerosol (SIA) consisting of ammonium nitrate and chloride, crustal soil, salt/dust, on-road traffic, non-crustal dust, and secondary sulfate. The ammonium nitrate/chloride factor was likely from coal-combustion similar to what had been seen in Beijing (Li et al., 2019a). The crustal soil and non-crustal dust are distinguished by the presence of trace elements in the non-crustal profile such that it would more commonly be named road dust. The salt/dust appears to be mostly fresh sea salt. Given the presentation, it is impossible to determine if there has been chloride displacement. The ammonium nitrate/chloride factor represented 40.9% of the fine PM mass with 33.7% being secondary sulfate. On-road traffic (10.8%), crustal soil (9.47%, and non-crustal soil (5.13%) made up the rest of the fine PM with the salt/dust attributed only to the coarse PM. There were no temporal patterns of the source contributions provided and thus, overall, the source apportionment information was sparse and uncertain.

Guangzhou

There are no reports of recent source apportionment studies based on composition data analysis. Li et al. (2020b) used combinations of methods of analysis and data analysis to apportion the $PM_{2.5}$ in Guangzhou. Samples were collected from October 16, 2013 to July 18, 2014 with every day sampling for one month selected from each season. The resulting 92 samples were analyzed for water soluble ions, elements, and OC/EC and used in a PMF analysis. Stable isotopes of oxygen and nitrogen were analyzed to apportion the NO^-_3 among coal combustion, traffic emissions, and biogenic sources using a Bayesian mixing model. Differences in apportionment of the NO^-_3 between these two methods were used to apportion it among the three sources such that after the secondary nitrate assignments, the six main sources of $PM_{2.5}$ were determined to be traffic emission (30.6%), biomass burning (23.1%), coal combustion (17.7%), ship emission (14.0%), biomass boiler (9.9%) and industrial emission (4.7%). Fossil/non-fossil source contributions to organic carbon (OC) and element carbon (EC) inferred from the ^{14}C measurements and compared with the corresponding results in the PMF model. These results suggested that the PMF analysis has underestimated the biogenic sources of OC but had provided an adequate apportionment of the EC. The final apportionment was biomass boiler (9.9%), ship emissions (14.0%), biomass burning (18.8%), traffic emissions (26.6%), coal combustion (14.6%), industrial emissions (4.8%), and secondary aerosol (11.3%). They then apportion the secondary aerosol mass to the actual source types and obtain a final apportionment of biomass boiler (9.9%), ship emissions (14.0%), biomass burning (23.1%), traffic emissions (30.6%), coal combustion (17.7%), industrial emissions (4.7%). Although these results are based on older samples, the combination of methods provides a useful example of how results can be refined through multiple measurements and data analytics. However, there are some uncertainties to their results. Ship emissions include SO_2 that will produce secondary aerosol. It is difficult to imagine that biomass boilers that are using solid fuels are not also going to emit secondary organic aerosol precursors.

Shenzhen

The only recent paper on source apportionment in Shenzhen is by Sun et al. (2019). They collected $PM_{2.5}$ filter samples at 5 sites in Shenzhen in March, June, September, and December 2014. The mean annual $PM_{2.5}$ concentration was 35.7 $\mu g/m^3$. Vehicular emissions, secondary sulfate, secondary organic aerosol (SOA), and secondary nitrate were the major sources, contributing 27%, 21 %, 12%, and l0%, respectively. Industrial emissions (8%), sea salt (3%), building dust, and coal combustion (7%) were quantified. Fugitive dust, biomass combustion, and ship emissions accounted for another 11% of the $PM_{2.5}$ mass.

Mongolia—Ulaanbaatar

Gunchin et al. (2019) presents the only recent apportionment of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ in Ulaanbaatar. Samples were collected with a Gent sampler (Hopke et al., 1997) on Mondays and Thursdays in 2014 to 2016 resulting in 184 pairs of polycarbonate filters for analysis. During high pollution periods in the winter, the sampler was operated on an on-off cycling scheme to avoid overloading and clogging the filter. Black carbon was measured with an EEL Smoke Stain Reflectometer (Model 43D) and elements via XRF. The resulting data were analyzed using EPA PMF V5.

The average $PM_{2.5}$ mass concentration was 92.8 ± 95.2 $\mu g/m^3$ while the $PM_{2.5\text{-}10}$ mass concentrations averaged 131.0 ± 102.0 $\mu g/m^3$. The sum of the measured $PM_{2.5}$ constituents represented 12.7% of the average mass and the reproduced mass (Malm et al., 1994) represented only 22.3%. An interesting feature of the composition data is a very high correlation between BC and S suggesting that the lump coal burning in stoves is a likely substantial source of primary sulfate as seen in the study of emissions from Chinese coal stoves (Dai et al., 2019) as well as likely having high sulfur content in their liquid fuels also producing primary sulfate (Santoso et al. 2008). A somewhat higher fraction of the $PM_{2.5\text{-}10}$ composition has been determined. However, the apportionments are based on low fractions of the composition being determined with nitrate and particularly organic carbon being the likely missing mass species. Thus, limited numbers of sources were identified (4 for each size cut) and do not apportion more mass than the reproduced mass values.

For $PM_{2.5}$, the resolved sources were Traffic (30.7%), Soil (33.1%), Coal Combustion (26.0%) and Oil Combustion (10.2%) with a total explained mass of 53.3 $\mu g/m^3$ or 45.6% of the average mass concentration. Sulfur appears in the Traffic, Coal, and Oil factor profiles, but it is not possible to know how important they were in each profile since the DISP intervals were not provided. For $PM_{2.5\text{-}10}$, the resolved sources were Traffic (41.9%), Soil (34.4%), Coal Combustion (15.6%) and Oil Combustion (8.1%) with a total explained mass of 53.3 $\mu g/m^3$ or 40.7% of the average mass concentration. However, there was no sulfur in either the Traffic or Oil profiles and that raises questions about their accurate identification. Overall, there are serious concerns about these results given the limited compositional analyses relative to the overall measure PM mass values.

Korea—Seoul

Korea has also been implementing regulatory controls to reduce air pollution and improve public health. In 2017, the government introduced the Action Plan on Fine Dust was implemented in 2017 whose goal is to reduce $PM_{2.5}$ emissions by 30% compared with the level in 2014 by 2022 and to reduce the annual average $PM_{2.5}$ concentration in Seoul to 17–18 $\mu g/m^3$ (Lee, 2018). Sampling/analysis of $PM_{2.5}$ was performed in Seoul using the same methods as used in Beijing by Park et al. (2022). In Seoul, samples were collected on 294 days in 2019 resulting in an average mass concentration of 27.1 ± 19.0 $\mu g/m^3$ with a seasonal maximum of 36.2 ± 19.7 $\mu g/m^3$ in winter and minimum in fall (18.6 ± 10.6 $\mu g/m^3$).

The compositional data were analyzed using dispersion normalized PMF and nine factors were resolved. They were: secondary nitrate (25.5%), secondary sulfate (20.5%), biomass burning (11.3%), incinerator (10.5%), mobile (10.0%), residual oil combustion (10.0%), coal combustion/industry (5.9%), soil (5.1%), and aged sea salt (1.4%). Decreased contributions from secondary sulfate and increased secondary nitrate were observed in Seoul compared to prior studies (Heo et al., 2009; Park et al., 2020). This result suggests that the reduction in sulfate allowed more ammonia to be available to form particulate nitrate (Kong et al., 2014).

A second recent study examined the role of local emissions versus transported $PM_{2.5}$ to the apportioned sources in Seoul by examining the differences between COVID-19 lockdown periods and other times when normal activities were occurring (Kim et al., 2022). In this case, hourly data were available from a set of semicontinuous instruments operated by the National Institute of Environmental Research (NIER) in Seoul. This supersite is in a residential and commercial complex located 6.3 km northwest of Seoul city center, adjacent to high traffic zones. Hourly values of elements, water-soluble ions, and OC/EC from January to March in 2019 and 2020 were available for the DN-PMF analyses. Ten sources were resolved: sulfate (25.9%), secondary nitrate (24.5%), biomass burning (21.3%), traffic (9.2%), mining industry (4.2%), district heating (3.8%), soil/road dust (2.5%), metallurgical industries (1.8%) and residual oil combustion (1.2%). These results are for winter/early spring only and thus, cannot be directly compared with those of Park et al. (2022) for a different location in Seoul. Substantial reductions in sulfate, nitrate, biomass burning, mining, district heating, and oil combustion were observed from 2019 to 2020. However, there were increases in vehicular emission and residential coal combustion. By examining a period with weak transport conditions and poor local dispersion, they could estimate the reductions in the various sources and separate those reductions between transported and local materials. The results are presented in Table 5.

Table 5. Reductions in contributions for transboundary and local sources during winter/early spring 2020 relative to winter/early spring 2019.

Source	Transboundary Impact ($\mu g/m^3$)	Local Impact ($\mu g/m^3$)
Secondary Nitrate	-28.86	-5.16
Sulfate	-21.91	-6.41
Biomass Burning	-16.90	0.02
Mining Industry	-4.84	-0.87
District Heating and Incineration	-2.53	-0.96
Soil	-1.78	0.32
Metallurgical Industry	-0.21	-0.23
Oil Combustion	-0.97	-2.41
Coal Combustion	0.31	1.96
Mobile	2.08	4.23

Thus, it will take the implementation of controls in both Korea and upwind countries such as China to substantially improve particulate air quality in Seoul.

Malaysia—Kuala Lumpur

Jamhari et al. (2022) reported the composition, concentrations, and source apportionments of size-fractionated urban PM in Kuala Lumpur, Malaysia. They collected multiple size samples on 55 mm quartz filters between February 17 and December 3, 2017 so as to cover the 4 seasons, the northeast monsoon (NE), the southwest monsoon (SW) and two intermonsoon seasons (IM1 and IM2). They employed the Nanosampler 3180 with effective cut-off diameters at <0.1, 0.1–0.5, 0.5–1, 1–2.5, 2.5–10 and >10 µm, which are represented by $PM_{<0.1}$, $PM_{0.1–0.5}$, $PM_{0.5–1.0}$, $PM_{1.0–2.5}$, $PM_{2.5–10}$, and $PM_{>10}$, respectively. They were analyzed for water-soluble ions by ion chromatography and for OC/EC by thermo-optical analysis. Elemental data were measured using ICP-MS. Sources were apportioned using EPA PMF V5. They used the OC/EC ratio method to separate primary (POC) and secondary (SOC) organic carbon (Turpin and Huntzicker, 1995) using $(OC/EC)_{primary}$ =2.0 as the basis of their analyses. Thus, they did estimate POC values that were greater than the measured OC. They also interpreted the carbon fractions, EC1 as char-EC and EC2 as soot-EC (Han et al., 2007). They combined the data to produce mass and compositional data for $PM_{2.5}$ that was then analyzed using EPA PMF V5.

They only resolved 5 sources based on their 72 samples; Sea Salt (18.6%), Mineral and Road Dust (4.7%), Biomass Burning and SIA (38.5%), Traffic Emissions (22.4%), and Industrial Emissions (15.9%). The inability to separate biomass burning normally indicated by high K+ and OC from ammonium sulfate and ammonium nitrate is surprising and suggests inexperience with developing the input data for PMF analysis although the limited number of samples also may restrict the resolution. Although they have carbon fraction data, it does not appear that neither OC/EC or the OC/EC fractions were used in the PMF analysis since they do not appear in the profiles presented in Figure 5. Thus, it appears they missed the opportunity to take full advantage of their data.

Singapore

There is a single paper on apportionment of 114 samples of $PM_{2.5}$ collected in Singapore from July 2015 to January 2016 (Yan et al., 2019). There are no details of when the samples were collected or the methods of analysis to produce the compositional data. They use their own non-negative least squares formalism, but with an improper weighting scheme. The advantage of using an explicit least squares formulation is to be able to provide individual weights to each data point to maximize the information content and minimize the influence of noise. They simply use the standard deviation of each sample. They also do not include the PM mass in the analysis and that along with proper weighting, provides a better apportionment of the PM mass. They could have done a regression analysis of the contribution values and the measured mass to get the apportionment, but they did not report that either. They identify the 8 sources as: Crustal material, biomass burning, shipping, sea salt 1, sea salt 2, waste incineration, abrasive/metallurgy, and vehicles. However, there is no information regarding the quantitative apportionment of the $PM_{2.5}$ mass.

Vietnam—Hanoi

Hien et al. (2021) reported the results of a source apportionment study conducted in a new urban area of Hanoi, Vietnam. Samples were collected in two size fraction using a dichotomous air sampler (Zambelli Twin Dust; Aquaria Tech s.r.l., Italy) that were set up to collect $PM_{1.0}$ and $PM_{2.5}$. Because of the high concentrations being sampled, the sampler was operated alternating between on and off (2 h on followed by 2 h off) throughout the 24-h period thereby providing a representative sample for that day. Samples were collected from November 2015 to June 2016 obtaining a total of 85 pairs. They used 0.4 µm pore polycarbonate filters that were then weighed and analyzed for chemical constituents. BC was measured with the EEL Smoke Stain Reflectometer. Following the BC measurement, the filters were cut into 2 equal pieces. One was leached with 18 MΩ water and analyzed for anions and cations with ion chromatography. Elements were determined using proton-induced X-ray excitation. EPA PMF V5 was used to perform the source identification and apportionment.

The mean (± standard deviation) 24-h mass concentrations of $PM_{1.0}$ and $PM_{2.5}$ were 30.1 ± 13.9 $\mu g/m^3$ and 44.5 ± 21.0 $\mu g/m^3$, respectively. They used SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}, BC, and a limited number of elements (Al, Fe, Si, Mg, K, Pb, Si, and Zn). They did not have a measure of organic carbon. They resolved 7 sources for both $PM_{1.0}$ and $PM_{2.5}$: road dust, coal, construction, sea salt, primary vehicular emissions, biomass burning, and long-range transported (primarily ammonium sulfate with some BC and Pb, particularly in $PM_{1.0}$). There is also a substantial amount of unexplained mass that is likely the OC. These data would be a good candidate for using unmeasured mass (Hopke et al. 2003) since there is relatively complete mass analysis except for OC. The apportionment results are summarized in Table 6.

Although they properly exclude S because they included sulfate, they used both K and K^+, resulting in double counting its influence (JRC, 2014). There are a number of issues with their factor assignments, but they again do not provide the DISP intervals to examine the degree of association a given species has with a specific factor. For example, their $PM_{2.5}$ sea salt has too much Cl^- for the amount of Na^+ and there is too little Mg and Ca. $PM_{2.5}$ construction has it most abundant constituent being Mg^{2+}, then Ca^{2+}, and Na^+ so Na^+ has been apportioned from sea salt to construction. They could use ratio constraints in EPA PMF to adjust these obvious issues when there is known stoichiometry such as for sea salt.

Table 6. Fractional source apportionment of $PM_{1.0}$ and $PM_{2.5}$ in Hanoi, Vietnam

Source	$PM_{1.0}$	$PM_{2.5}$
Coal	15.4%	3.0%
Vehicle	10.7%	8.0%
Road	5.5%	26.9%
Sea Salt	5.1%	7.0%
Biomass Burning	4.3%	9.1%
Construction	1.0%	4.0%
Long-Range Transport	32.2%	24.1%
Unexplained	25.7%	17.9%

Philippines—Manila

Tseng et al. (2021) performed source apportionments at both Kaohsiung Harbor in Taiwan and Manila Harbor in the Philippines. Samples of $PM_{2.5}$ were collected on 7 consecutive days in each of 4 seasons from May 2018 to January 2019. The quartz filters were divided into quarters and analyzed for water-soluble ions, metallic elements, carbonaceous constituents, anhydrosugars, and organic acids. They used both CMB and PMF to provide source apportionments and compared the constituent and source contributions between the two harbor areas. In Manila, they resolved 6 factors. However, they were unable to provide clear assignments of sources that these factors represent. The factors were designated as: Road dust/coal-fired boilers (17.4%); Ship emissions/vehicular exhaust (19.1%); Industries (17.7%); Vehicular exhaust (12.6%); Secondary aerosol/soil dust/biomass burning (21.3%); and Ocean spray/biomass burning (11.8%). They did not use the DISP intervals to identify which species have small or large rotational ambiguity and thus, had substantial or little importance in identifying the source type, respectively. Although they had molecular marker data, the very limited number of samples precluded clear source resolution. Secondary inorganic species were distributed over multiple profiles without clear sulfate and nitrate factors. This result suggests they used too small uncertainties and that results in distributing species in multiple profiles to meet the requirement of fitting the data to too tight requirements. The comparisons between sites were done using t-tests that are very likely inappropriate since environmental data are rarely normally distributed and that is a requirement for calculating the probabilities that the data distributions between the sites were similar. The application of CMB is questionable in Manila since there is no indication that local source profiles were measured as part of this project. Thus, the source apportionments provided in this work have high uncertainty.

Pabroa et al. (2022) collected samples at 3 sites in Manila (NAMRIA, MMDA EEDSA, and Valenzuela) on Wednesday and Sunday from June to November 2016 using a Gent sampler (Hopke et al., 1997) thereby providing at least 75 fine (≤ 2.5 μm) and coarse (2.5 to ≤ 10 μm) samples per site. Black carbon (BC) was determined using an M34D Smoke Stain Reflectometer and elements were measured using proton-induced X-ray excitation. The source apportionment was performed using PMF as implemented in PMF2 (Paatero, 1997). PMF2 is an older approach to applying an explicit least-squares method that uses a different technique for the non-negativity constraints and it does not provide meaningful error estimates. The partial year average $PM_{2.5}$ mass concentrations ranged from 19.0 (4.8-41.4) to 25.9 (5.7-74.2) for the 3 sites. A notable feature of the compositions in Manila is the high BC concentrations constituting 31–46% of the $PM_{2.5}$. Across South and Southeastern Asia, Manila and Dhaka, Bangladesh had comparable concentrations (~8.9 $\mu g/m^3$) that was only exceeded by Colombo, Sri Lanka. However, this study had no direct measure of organic carbon or nitrate and thus, sources that depend on these species would not have been identified and their associated mass concentrations will be partitioned to other source types. The apportionments at the three sites are summarized in Table 7.

Table 7. Fractional source apportionments of $PM_{2.5}$ at the 3 sites in Manila

Source	Valenzuela	MMDA	NAMRIA
Soil	9.2	11.0	13.5
Vehicular emissions	35.2	20.0	32.5
Sea salt	8.5	13.3	14.5
Industry	14.1	30.7	6.7
Smoke (biomass)	27.6		24.9
Secondary sulfate		21.2	7.9
Industry (Zinc)		3.9	
Industry (Zinc-Lead)	5.4		

As can be seen from Table 6, a total of 8 sources were identified, but only 6 at any given site. There are a number of questions regarding these results. There is a significant port in Manila Harbor and thus, the lack of marine diesel (a Ni-V factor) is surprising. The high variability in secondary sulfate across the city is also difficult to understand since it would be anticipated that coal-fired power plant SO_2 emissions would have been oxidized to secondary sulfate and that aerosol would be distributed across the region. Depending on the sulfur content of their diesel fuel, there would be primary sulfate associated with heavy diesel traffic areas (Santoso et al., 2008). These data would have been better analyzed using a calculation of the unmeasured mass (Hopke et al., 2003) to provide input data that fully reproduces the measured mass values. These data only represent 6 months of data so the representativeness of these results are uncertain.

Indonesia—Jakarta

In Jakarta (Vital Strategies, 2022), $PM_{2.5}$ filter samples were collected at 3 sites, Gelora Bung Karno (GBK), Kebon Jeruk (KJ) and Lubang Buaya (LB), that were selected based on land use features, weather, and other considerations to capture potential variation in air pollution sources. Sampling occurred during a wet season (October 2018 to March 2019) and a dry season (July to September 2019). Samples were not collected on rainy days and the total numbers of samples are not provided. At each site, 2 Air Metrics Mini-Vol samplers (Baldauf et al., 2001) were deployed with one collecting on a 47 mm Teflon filter and the other on a 47 mm Quartz filter. BC was measured with an EEL Smoke Stain Reflectometer (Model 43D), elements by XRF, water-soluble ions by ion chromatography, OC/EC by thermal optical transmittance. They employed CMB analysis with 11 known source profiles with 6 to 9 sources being resolved at the 3 sites. The average daily $PM_{2.5}$ concentrations over the entire sampling period range from 39 $\mu g/m^3$ in central Jakarta (GBK) to 56 $\mu g/m^3$ in western Jakarta (LB). The results of their source apportionments are provided in Table 8.

Table 8. Source apportionment of $PM_{2.5}$ at 3 sites in Jakarta, Indonesia

	Wet			Dry		
Source	GBK	KJ	LB	GBK	KJ	LB
Unidentified	31	26	22	17	0	10
Secondary Aerosol	11	16	6	7	2	1
Vehicular Exhaust	41	32	38	42	57	43

Gasoline	19	7	13	18	28	35
Diesel	22	26	24	24	28	18
Non-Vehicular Primary	17	25	34	34	42	46
Paved road dust	6	1	0	0	9	0
Construction	0	13	0	0	0	0
Soil	1	0	1	12	10	18
Open Burning	0	11	11	0		9
Sea salt	10	0	9	22	22	19
Coal Combustion	0	0	14	0	0	0

The nature of the profiles and their origins are not provided. Given the reference to the US EPA's CMB8 software, it is likely that the profiles were obtained from the EPA's SPECIATE data base that provide profiles measured in the United States. For primary source profiles, the most recent one in that data base is from 1993. There are many profiles from individual vehicles or small groups of vehicles, but as previously noted, Subramanian et al. (2006) demonstrated the difficulties in utilizing the limited measured profiles to represent the fleet-average compositions. The wide spatial and temporal variations in the number and contribution of sources seems unrealistic. It would be interesting to reanalyze these data with PMF.

Bangladesh—Dhaka

The most recent full apportionment of $PM_{2.5}$ was reported by Begum and Hopke (2019). They analyzed the elemental and BC concentration data from samples collected with a Gent sampler (Hopke et al., 1997) from 1997 to 2015. They identified 8 sources (biomass burning, soil dust, brick kilns, fugitive lead, road dust, galvanizing plant, motor vehicles, and sea salt. They were able to relate some of the variations in contributions to air quality management policy changes.

To provide exposure data for an epidemiological study of cardiovascular morbidity and mortality (Rahman et al. 2021) and respiratory emergency department visits (Rahman et al., 2022), additional samples from the same location collected in the same manner were analyzed for elements by XRF in order to extend the data set through 2017 (Rahman et al., 2020). Rahman et al. (2020) then did a simplistic source apportionment between biomass burning, fossil fuel burning, and all other sources of $PM_{2.5}$. The mean source contributions varied by season: During the monsoon season, $PM_{2.5}$ was dominated by fossil-fuels sources (44.3%), whereas it was dominated by biomass-burning (41.4%) during the remainder of the year.

India—Delhi/New Delhi

Delhi/New Delhi has become one of the most polluted cities in the world with particularly notable problems in the late fall and winter in large part due to agricultural burning across the Indo-Gangetic Plain and the use of solid biofuels for residential heating in the surrounding areas. There are specific times such as the Diwali Festival at which air quality is degraded by extensive use of fireworks (Manchanda et al., 2022). However, their study only covered 12 days so it does not provide an apportionment that can be applied to a broader time period.

Shivani Gadi et al. (2019) sampled $PM_{2.5}$ on quartz filters in low volume samplers at 3 sites, twice or thrice a week during the period of December, 2016 to December, 2017. They analyzed the samples for OC/EC, water-soluble inorganic ions, and fifty-five organic species in six classes of organic compounds: n-alkanes (C11-C35), isoprenoid hydrocarbons (pristane and phytane), hopanes, steranes, polycyclic aromatic hydrocarbons (PAHs), phthalates, n-alkanoic acids (even numbered C12-C20), and levoglucosan. No elemental data were obtained and thus, constituents like road dust could not be resolved. The application of PMF resolved vehicular emissions (34.6%), biomass burning (26.8%), cooking emissions (15.7%), plastic and waste burning (13.5%) and secondary organic carbon (9.5%).

Jain et al. (2020) collected $PM_{2.5}$ and PM_{10} samples at the CSIR National Physical Laboratory, New Delhi, India, from January 2013 to December 2016 at least 2 times a week on pre-combusted quartz filters. There were 350 pairs of $PM_{2.5}$ and PM_{10} samples collected and then analyzed for elements using Wavelength Dispersive X-Ray Fluorescence, for OC/EC using a DRI2001 Thermo-optical analyzer, and water-soluble ions using ion chromatography. PMF analyses for both size cuts were performed using EPA PMF V5.

The four-year average concentrations of $PM_{2.5}$ and PM_{10} were 131 ± 79 $\mu g/m^3$ and 238 ± 106 $\mu g/m^3$, respectively. PM concentrations increased year-over-year from 2013 to 2016. Maximum monthly $PM_{2.5}$ concentrations were observed during the post monsoon season (October to December) while minimum values were found during the monsoon season. The source apportionment results resolved 8 source types: secondary nitrate (SN), secondary sulfate (SS), vehicular emissions (VE), biomass burning (BB), soil dust (SD), fossil fuel combustion (FFC), sodium and magnesium salt (SS), and industrial emissions (IE). The average annual contributions to $PM_{2.5}$ were: SN 11.7%; SS 9.0%; VE 16%; BB 23%; SD 13%; FFC 10%; Salt 6%; IE 10.3% and for PM_{10}, they were: SN 11.8%; SS 8.0%; VE 21%; BB 19%; SD 17%; FFC 7%; Salt 7%; IE 9.9%.

India—Mumbai

There is only one publication available for Mumbai. Police et al. (2018) reported the source apportionment of $PM_{2.5}$ and $PM_{2.5-10}$ collected in Trombay, Mumbai in 2010 and 2011using a Gent sampler (Hopke et al., 1997). Samples were collected twice weekly over the 2 years period. Elements were measured using X-ray fluorescence and BC was measured using an EEL Smoke Stain Reflectometer (Model 43D). Trombay is a heavily industrialized part of Mumbai with a number of identified industrial sources including major ports. For 2010, the average concentrations were: 31.80, 67.90. and 99.73 $\mu g/m^3$ for $PM_{2.5}$, $PM_{2.5-10}$, and PM_{10}, respectively, while in 2011, the average values were: 27.70, 52.40, and 80.70 $\mu g/m^3$.

They resolved 6 sources from each size fraction's data using EPA PMF V5 that they identified as: Fuel oil combustion, Coal/biomass combustion, Road traffic, Sea salt spray, Crustal material, and Metal industry. The "Fuel oil" is residual oil coming substantially from the marine diesel engine emissions at the ports. They observed fresh sea salt with no indication of substantial chloride displacement. The Metal industry was notable for high Cr and Mn, but relatively low Fe and some contributions to Cu and Zn. The apportionments for the 2 size fractions are summarized in Table 9.

Table 9. Fractional source apportionment for Mumbai (%) (Police et al., 2018).

Source Type	$PM_{2.5}$	$PM_{2.5-10}$
Crustal Material	8.7	8.7
Sea Salt Spray	6.1	15.0
Coal/Biomass Combustion	25.5	13.8
Road Traffic	17.7	12.6
Fuel Oil Combustion	19.0	11.2
Metal Industry	10.6	7.9
Unknown	12.4	15.0

Their analyses did not provide any measure of OC or ammonium nitrate. They did not provide any of the diagnostics that are provided by EPA PMF V5 and did not apply any of the meteorological methods like conditional bivariate probability function and potential source contribution function that would be useful in identifying the likely source directions and locations.

Pakistan—Karachi

There are two relatively recent publications, Shahid et al. (2018) and Lurie et al. (2019), that report source apportionments for PM_{10} and $PM_{2.5}$, respectively. However, the Shahid et al. study analyzed data from 9 samples collected in March and April 2009 and the Lurie et al. study examined samples collected from August 2008 through August 2009. Thus, there are no studies of the recent composition or apportioned PM sources for Karachi.

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