Positive Matrix Factorization on Source Apportionment for Typical Pollutants in Different Environmental Media: A review

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Abstract

A bibliometric analysis of published papers with the key words "positive matrix factorization" and "source apportionment" in 'Web of Science', reveals that more than 1000 papers are associated with this research and that approximately 50% of these were

produced in Asia. As a receptor-based model, positive matrix factorization (PMF) has been widely used for source apportionment of various environmental pollutants, such as persistent organic pollutants (POPs), heavy metals, volatile organic compounds (VOCs) as well as inorganic cations and anions in the last decade. In this review, based on the papers mainly from 2008 to 2018 that focused on source apportionment of pollutants in different environmental media, we provide a comparison and summary of the source categories of typical environmental pollutants, with a special focus on polycyclic aromatic hydrocarbons (PAHs), apportioned using PMF. Based on the statistical average, coal combustion and vehicular emission, are shown to be the two most common sources of PAHs, and contribute much more to emissions than other sources, such as biomass burning, biogenic sources and waste incineration. Heavy metals were mainly from agricultural activities, industrial and vehicular emissions, and mining activities.. Quantitative source apportionment on pollutants such as VOCs and particulate matter were also apportioned, showing a prominent contribution from fossil-fuel combustion. We conclude that, aside from natural sources, abatement strategies should be focused on changes in energy structure and industrial activities, especially in China. Source apportionment of typical POPs including polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), halogenated flame retardants (HFRs) and perfluorinated compounds (PFCs) are less comprehensive and further study is required.

environmental media

1 Introduction

Due to rapid global industrialization and urbanization, both the scale of emission and number of species of anthropogenic pollutants released into environment have increased in the past decades. For example, it was estimated that there were approximately 2749 Gg organic carbon (OC) compounds and 1857 Gg elemental carbon (EC) pollutants emitted in China in 2012. Compared with those emitted in 2000, the OC emissions have increased by 29% (from 2127 to 2749 Gg) and by 37% for EC (from 1356 to 1857 Gg). [1] Furthermore, inputs of heavy metals and metalloids, such as Cd, As, Hg, Pb and Cu from pollution sources such as industrial atmospheric emissions, livestock manures, fertilizers, and sewage irrigation to agricultural soils ranged from 125 to 78990 tons/year in China[2] between 2008 and 2018 These pollutants, are now largely considered ubiquitous in environment media, imposing potential detrimental effects on human and environmental health. Providing reliable information on the sources of these pollutants is therefore important in order to establish effective abatement strategies for environmental pollution prevention. To understand these sources, researchers have used various receptor models to apportion source categories and positive matrix factorization (PMF) is one of the most widely employed. Other receptor models include chemical mass balance (CMB), principle component analysis (PCA), and UNMIX (a multivariate receptor model). Compared with these quantitative receptor models, PMF can give a corresponding weight to the "uncertainty" of the data

and include this information in the final result. Data that have missed or that are below the detection limit are accorded greater uncertainty to reduce the interference of these data on the source resolution while non-negative constraints on factor loading and factor scores make the results more reasonable (http://www.epa.gov/heasd/products/pmf).

The application of these models to source apportionment studies has been extensive. For example, Sofowote et al., (2008) used PMF and PCA with multiple linear regression analysis (MLR) to determine the sources of polycyclic aromatic hydrocarbons (PAHs) in suspended sediments in Hamilton Harbour, Canada. The results showed that both methods identified vehicular emission, but that PMF was able to further differentiate this source into gasoline and diesel emissions s,[3] Cao, et al., (2011) analyzed the PAHs in surface sediments of Shantou coastal mangrove zone, China, and compared the apportionment results of PMF with those obtained by PCA and MLR, and also found that PMF apportioned more distinct source categories than the other method.[4] Khairy, et al., (2013) apportioned the sources of PAHs in the aerosol of Alexandria, Egypt andalso concluded that the PMF model afforded better source identification than factor analysis (FA) with MLR.[5] The more reliable source apportionment by PMF is a significant advantage as PMF factor loadings are statistically scaled according to their relative contributions.[6] However, now that there have been a large number of studies that have applied PMF to apportion sources of pollutants in various environmental media there is a need to evaluate the approach. This review aims to assess the source categorization by PMF using results from published

papers and to determine potential research directions on source apportionment using the technique. It therefore provides important information for policy makers to establish targetted abatement strategies for environmental pollution emissions.

2 PMF model

Paatero et al., (1993) developed the PMF method for particle source analysis based on traditional factor analysis (FA).[7] Since then, PMF has been widely used for source identification of anthropogenic pollutants in different media such as air, water and soil. As a receptor model, PMF could run with non-negative constraints that define the concentration matrix of chemical species measured at receptor sites, as the product of source composition and contribution factor matrices with a residue matrix. The model principle is briefly explained as follows:

$$X_{ij} = \sum_{k=1}^{p} A_{ik} F_{kj} + R_{ij}$$

where X_{ij} is the concentration of species i measured in the sample j; F_{kj} is the contribution of the source k to the sample j; A_{ik} is the concentration of the species i from the source k; and R_{ij} is the residual for each sample/species.

The task of PMF is to minimize the sum of squares Q defined as:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{X_{ij} - \sum_{k=1}^{p} A_{ik} F_{kj}}{S_{ij}} \right)^2$$

The robust Q value reduces the impact of outliers in the fitting of the model. The theoretical Q value should be approximately equal to the number of data entries in the

concentration file for a model. The uncertainties for each sample are calculated using measurement uncertainties (MU%) and method detection limits (MDL). If the concentration ≤ MDL, the uncertainty S_{ij} is calculated as:

$$S_{ij} = \frac{5}{6} \times MDL$$

When the concentration > MDL, S_{ij} is calculated as:[8]

$$S_{ij} = \sqrt{(MU \times concentration)^2 + (MDL)^2}$$

The MDL refers to the minimum concentration of the analyte being detected by a system at a reasonable confidence level under certain conditions. In general, the detection limit is calculated by analyzing a blank sample or a standard sample containing a low concentration, calculating the standard deviation of the analysis result, and determining the standard deviation multiple. The formula is as follows:

$$MDL = t(n-1, 1-\alpha=0.99) \times S$$

Where t is a unilateral 99% confidence interval for Student's t-distribution with $n-1$ degrees of freedom; n is the number of repetitions; S is the standard deviation or noise of the measurement.

Importantly, a critical step for PMF analysis is to determine the number of factors. A number of factors between 3 and 7 are examined with the optimal number of factors determined from the slope of the Q value versus the number of factors. For each run, the stability and reliability of the output are checked based on the Q value, residual analysis and correlation coefficients between observed and predicted concentrations. Finally, the optimum factor solution, which gives the most stable results and easily interpretable factors compared with the source factors apportioned by previous similar

studies, are chosen.

3 PMF on source apportionment for pollutants

PMF have been used for the source apportionment of a number of enviromental pollutants, especially POPs, PAHs, heavy metals and volatile organic compounds (VOCs).[9-16] However, over the last decade there have been many studies on the sources of particulate chemical composition from filter sampling, source resolution of particle number concentration (PNC) and particle number size distribution (PNSD) using PMF [17-21] Figure 1 shows the application frequency of PMF on source apportionment for PAHs, heavy metals, VOCs and PNC. It can be seen that the f frequency of PMF application for these pollutants has increased sharply, demonstrating the increasing popularity of the PMF method. Figure 2 shows a concept map of source categories appotioned using PMF according to pollutant types. Based on this concept map, Figure 3 shows the stacked bar ratio of the source apportionment forPAHs, heavy metals, VOCs and PNC from 2002 to 2018 both globally and for China using PMF. It is clear that, aside from PNC, the other studies were mostly distributed in Asia contributing, on average, 61.2% of the total. From a statistical analysis of the avaliable papers in "Web of Science", we found that over 90% of the studies in Asia were conducted in China and as a result the distribution characteristics for China are also analyzed and presented in Figure 3b. In recent years, China has suffered from extensive air pollution, especially in megacities such as Beijing and Shanghai. The high frequency of PMF applications on PAHs, heavy metals and VOCs in China is possibly due to the

environmental pollution associated with rapidly expanding economic and industrial developments. By contrast, the United States has experienced significant emissions reductions in the last two decades leading to a decreases in $PM_{2.5}$ and major gaseous pollutants (SO_2, CO, NO_x). Howeverit is now recognised that, compared with these pollutants, submicron particles (SMPs, <1 μm) and ultrafine particles (UFPs, <100 nm) are more directly related with adverse effects upon human health. As there are limited data for the sources of ultrafine particles, PNC studies in the USA have been relatively high over the last decade. Combining PNC and chemical composition information for PMF source analysis may provide a better means by which to analyze the true source of particulate matter, providing more data to establish effective pollution prevention strategies. In the following sections, the studies of PAHs, heavy metals, VOCs and PNC will be presented in more detail and further discussed according to their source categories.

3.1 PAHs

As a family of organic compounds that consist of fused aromatic rings, PAHs are known to cause carcinogenic and mutagenic effects on humans.[9] Sixteen species of PAHs have been listed as priority pollutants by the United States Environmental Protection Agency (USEPA) and seven of these are classified as possible human carcinogens by International Agency for Research on Cancer (IARC). As they mainly originate from anthropogenic activity, such as domestic and industrial coal combustion, biomass burning and vehicle emission, PAHs, once emitted, are ubiquitous in the

atmosphere. Thereafter, they may be transferred to surfaces via atmospheric deposition, where they may be adsorbed by organic matter in soils, resulting in a long retention time in the environment.[16] Understanding the sources of PAHs is of great importance for reducing human exposure to these toxic pollutants and preventing the environmental pollution associated with them.

3.1.1 Solid phases

Figure 4 summarizes the source categories and their contribution of PAHs apportioned by PMF in solid phases, such as soil, sediment and street dust from background and urban areas of Asia. It may be seen that these studies were mostly conducted in China and that the sources of these PAHs are mainly derived from various anthropogenic combustion processes, including coal combustion, vehicular emission, petroleum, coke and biomass burning. Non-combustion sources include the use of preservatives such as creosote as well as natural activities, including air-soil exchange and biogenic origins. In urban areas, the source apportionment by PMF analysis indicated that, based on yearly averages, traffic emission and coal combustion contributed more than 50% of the PAHs and that source categories were regionally distinct with strong seasonal variations. For example, the main sources of PAHs in soils from Dalian, northeastern China in the summer were coal combustion (46%), diesel engine (30%) and gasoline engine (24%) emissions; while in winter, they were from coal-fired boilers (72%), general vehicular emissions(20%) and, more specifically, gasoline engines (8%).[22] With strong seasonality of these source categories, this study

indicated that coal combustion in winter and traffic exhaust in the summer, dominated the sources of PAHs in these soils . Similarly, in Urumqi, China, coal combustion was also found to contribute approximately 50% of PAHs in the soil.[23] However, unlike Dalian and Urumqi, the PAHs in the soils of urban Shanghai were mostly associated with highly developed industrial activity and the rapid increase of vehicle number. Based on the PMF analysis, there were four emission sources, namely, unburned petroleum, creosote and coke tar usage and vehicle emissions, which were identified as contributing over 60% of the total PAHs.[24] Similar source categories and a higher relative contribution from industrial and vehiclar sources (over 80%) were found in Rizhao city of Shandong Province, China.[25] As regards wetland soils and lake sediments, such as wetlands ofin the Liao River and Lake Guchenghu, China, sources from coal combustion, biomass burning and traffic emissions contributed up to 80% of the PAHs.[26-27] For urban soils   in cities such as Nanjing, Xinzhou, and Daqing in China, coal, biomass, and oil combustion dominated.[28-30] These results suggest that the source categories and their contribution to PAHs in urban soils varied significantly across the cities, especially between the south and north China. , Changes   to energy structures are likely to be required for the abatement of PAHs in these locations.

Beyond China, PMF modeling   also identifies differences in PAH sources . For example, at Ulsan, Korea, industrial emissions and vehicle, especially diesel usage, contributed over 80% of the PAHs in soil, [31-32] while in the Gulf of Thailand, the contributions of PAHs from petroleum residue and air–water exchange was estimated to be 44% and 25% respectively, owing to direct land-based inputs (large amounts of

sewage, runoff, and sediments), gas absorption and the topography of the semi-enclosed tropical marine embayment.[33] Along the Jhelum riverine system of the Lesser Himalayan Region in Pakistan, biomass combustion and vehicular emissions via long range atmospheric transportation from neighboring countries were revealed as important sources of the PAHs.[34] The high contribution of sources from anthropogenic combustion in urban, coastal and "pristine" regions suggests that the accumulation of PAHs in soils has been elevated not only by urbanization but also poses potential environmental risks in areas remote from cities and industrial sources

3.1.2 Air

Figure 5 provides a comparison on the source categories and their contribution to PAHs in the air (gas or particle phase) apportioned by PMF with other studies at various sites worldwide. The sources of PAHs in the air are mainly dominated by anthropogenic activities, such as coal combustion, biomass burning and vehicular emission, with a very minor contribution from natural processes. As early as 2003, Larsen and colleagues identified four sources of PAHs, vehicle (16%), coal (36%), oil (15%), wood (35%), in the urban atmosphere of Maryland, USA using PMF[9] This study was the first to quantitatively apportion a dominant contribution of coal and wood combustion (over 70%) to ambient PAHs. Then, in 2004, Lee et al., (2004) found a source from "air-surface exchange" for atmospheric PAHs (gas + particle) and showed that this contribution could be as high as 25% of the total source categories. The "exchange" here, indicates a possible re-emission of aged PAHs from contaminated soil or

volatilization of PAHs directly from the water column of the Hudson River Estuary to the atmosphere. These PAHs are first emitted in the vapor phase and later absorbed by particulates.[35] In 2010, two northern Chinese cities, Harbin and Xi'an, were used to identify the sources of atmospheric PAHs during different seasons.[36,37] As expected, coal combustion was the largest contributor of PAHs in the 'heating season'; while in the 'non-heating season', contributions were mainly from vehiclar emissions. Technically, coal-fired centralized heating is provided due to the space heating policy in northern China, while this is not allowed in the south. Even so, there still were data that showed a dominant contribution from coal utilization in the south as demonstrated by the results that coal burning (30.5%) and gasoline engine emission (29.0%) were the two major sources of PAHs in the $PM_{2.5}$ in Shanghai, followed by diesel engine emission (17.5%), air-surface exchange (11.9%) and biomass burning (11.1%).[38] Similar to Harbin and Xi'an, there were also distinct seasonal variations of source categories in Shanghai. According to Wang et al., (2015), the highest source contributor for PAHs in fall and winter in Shanghai was coal combustion (30.5%) and vehicular emission (34.5%), respectively; while in spring and summer, air-surface exchange contributed the most (27.1% and 59.5%, respectively).[38] Ambient temperature and long range transport driven by the east Asian monsoon were both factors affecting the variation in source categories.[39,40] However, for countries beyond China, such as Egypt[5], Spain[41], Turkey[42], Iran[43], Malaysia[44] and Nepal[45], the sources of PAHs in the atmosphere were generally associated with petroleum usage including gasoline/diesel emission, natural gas combustion and stationary sources from industrial activity.

Between 2014 and 2019, further data on atmospheric PAH sources have been reported in various urban areas, including China[38-40], India[46,47] and Iran[48-50] as summarized in part of Figure 6. It may be concluded that the common sources of atmospheric PAHs in these regions have been relatively stable over the past 10 years although contribution ratios have fluctuated. Considering the similar source categories of PAHs in both solid phases (Figure 5) and aerosols (Figure 6), it is reasonable to suggest that only strict control of emissions will effectively reduce PAHs occurrence in the environment.

3.1.3 Water

Due to their low solubility, PAH concentrations in water often low and close to the detection limit, resulting in great uncertainty in source apportionment using PMF. Thus, researchers have usually integrated the sediment and water and used PAH sources in the sediment to indicate those source categories in the dissolved phase (DP) and suspended particulate matter (SPM). For example, Yang, et al., (2013) detected the PAHs in the topsoil, water, and sediment along the Jinjiang River entering Quanzhou Bay, China and apportioned three source categories: coal combustion (39.6%), traffic emission (27.3%) and wood/biomass combustion (33.1%) for soil, while for sediment they were 34.5%, 29.2% and 36.3%, respectively. High correlation between the PAHs in DP and sediments indicated similar sources.[51] Qin, et al., (2014) investigated the concentrations of PAHs in the water-SPM-sediment system of Lake Chaohu, China. The results showed the sources of PAHs in sediment, apportioned by PMF, were mainly from coal combustion (40.5%), vehicle emission (38.5%) and biomass combustions (21.0%). Positive correlations between PAHs in the water and dissolved inorganic

carbon (DIC) indicated that the PAH distributions in the water were mainly controlled by the DIC, which might be attributed to PAH inputs from the surface runoff from farm land, and asphalt roads following strong precipitation events in the summer.[52] Zeng et al., (2018) detected PAHs in the overlying water and surface sediments of Gucheng lake in Nanjing, China. The results showed that PAHs in water and sediment correlated significantly. Diagnostic ratios and PMF indicated a strong influence of pyrogenic sources, principally biomass combustion and vehicle emission for PAHs.[53] Bi et al., (2018) investigated the concentrations and distribution of PAHs in surface water, SPM and sediments among areas under different intensive land uses (industrial areas, agricultural areas, inner city, suburban towns and island areas) in the Shanghai river network, East China. They used isomer ratios and PMF to apportion sources and found biomass and coal combustion contributed strongly to total PAHs, followed by natural gas combustion for the water and SPM, and vehicular emissions in the sediments, respectively. Vehicular emissions were the strongest contributors to the SPM and sediments of the inner city, indicating the strong influence of vehicular transportation to PAH pollution in the urbanized river network.[54] These studies demonstrate the spatial and temporal variability in the concentration, composition, and possible sources of PAHs in surface waters. High-resolution studies, both on remote lakes and urban rivers, are required to provide additional insight into the contamination and transport mechanisms of PAHs through the water column and into sediments

3.2 Heavy metals

Heavy metals enter into the surrounding environment via the weathering of rocks and minerals, and emissions from many human activities such as fossil-fuel combustion, mining, electroplating, smelting and other industrial processes..[55] Studies have shown that heavy metals may   transform from solid to either ionic or, through biomethylation, to organometallic forms, which easily migrate in air and water, posing a risk to the ecological quality of surface waters, as well as   to the health of animals and humans.[56,57] Due to their potential toxiccharacteristics, heavy metals and metalloids such as Cr, Cd, Hg, Pb, Cu and As have been listed as priory control pollutants by USEPA.[58,59] There have been hundreds of studies on source apportionment of heavy metals in soils using PMF in the past decades. Here, we focus on the source categories of heavy metals in the soil/road dust apportioned by PMF based on Chinese studies published in 2018 as presented in Table 1. In urban areas, such as Nanjing[60], Tai'an[61] and Tianjin[62], agricultural activities (i.e., fertilizer application) were found to be an important source (~approximately 30%) of heavy metal accumulation in the soils. In cities, including Xi'an[63], Baotou[64] and Changsha[65], natural emission was identified as a separate source category that contributed 20% of the heavy metals in the soil. Road dust from urban areas in Beijing and mining areas in Baotou of northern China were also distinct sources. In Beijing, four categories: traffic exhaust emissions (32.2%), fuel combustion (29.0%), manufacturing and use of metallic substances (24.8%), pesticides, fertilizers and medical devices (14.0%), were determined by PMF. Traffic exhaust emissions were the most important source in summer, while fuel combustion contributed most significantly to the total heavy metal concentration in winter.[66] In

Baotou, Inner Mongolia, mining (23%), asphalt (17%), vehiclar (23%) and soil sources (32%) were found to   influencing the level, patterns, and paths of heavy metals in road dust from areas surrounding mines..[67] However, their influence was weakened by distance, as the particles were mainly deposited via wind transport from the mining region. Even for the soils of remote areas, such as Hexi Corridor in northwest China[68], Yulin in Shaanxi[69], and Wengyuan in Guangdong[70], the contribution from anthropogenic activities to the accumulation of heavy metals in soils was significantly higher than those of natural sources, and this was verified by the results of cluster analysis. Therefore aside from natural sources, heavy metals in soil/road dust across China are mainly derived from agricultural activities, industrial and , traffic emissions and mining activities. These results provide a reliable and robust approach for heavy metals source apportionment in these areas with a clear potential for future application in other regions across China. However, more detailed qualitative source categorisation combined with quantitative source apportionment of heavy metals are needed to provide data for control and reduction of heavy metal inputs to soils.

It should be noted that a focus on 'redox-active' metals are important in terms of health effects. By inducing oxidative stress through generating reactive oxygen species (ROS), these metals, for example, Fe, Cr, Cu, and Mn maylead to inflammation of target cells and organs.[71] By virtue of novel semi-continuous metal monitoring and   time-resolved source apportionment using PMF, a study in central Los Angeles reported that contemporary traffic contributed considerably to Fe and Cu in the urban atmosphere? and soil/road dust was the major source of Mn and Cu. Secondary aerosol was found to

make a very minor contribution to concentrations. It is important to highlight that traffic has a major impact on all redox-active metals. Moreover, contemporary traffic contributes more to metals in the cold season, while in the warm season, soil/road dust contributes more.[72] Hence, studies on the sources, transport, and temporal-spatial characteristic of redox-active metals are significant since they provide direct information on the abatement of human exposure.

3.3 VOCs

A number of VOC species such as benzene and ethylbenzene are carcinogenic and toxic and so can affect human health both directly and indirectly.[73] As important precursors of ozone and secondary organic aerosol,[74] VOCs are ubiquitous in air and emitted from a wide variety of natural sources, such as foliar emissions and wildfires, as well as anthropogenic sources such as fuel combustion, vehicle exhaust, industrial emissions and paints/solvent usage.[75-77] Table 2 provides a comparison on the source categories of VOCs in the atmosphere apportioned by PMF in 2018. For example, Bari et al., (2018) investigated concentrations of 38 VOCs in the urban area of Calgary, Canada and found the most dominant compounds were alkanes (58%), followed by halogenated VOCs (22%) and aromatics (11%). PMF revealed 9 VOC sources, namely oil/natural gas extraction/combustion (26%), fuel combustion (20%), traffic sources including gasoline exhaust, diesel exhaust and mixed fugitive emissions (10-15%), industrial coatings/solvents (12%), dry cleaning (3.3%) and biogenic sources (3.5%) as well as a 'background' source (18%).[78] In a heavy oil extraction area in Alberta, Canada,

Aklilu et al., (2018), used PMF to apportion three tracer species: acetone (19%), benzene/hexane (7%), toluene/xylene (5%), and two factors: regional air mass (30%), oil evaporation (15%), for the elevated concentrations of 22 hydrocarbon VOCs.[79]

In China, studies to apportion VOC sources in areas including oil fields and industrial parks, showed source categories associated with technical uses (i.e., oil refining processes, asphalt and foundry production)[80-81] while in urban areas, such as Taiwan[82], Nantong[83], Beijing[84] and Wuhan[85], many and various anthropogenic sources, including industrial processes, solvent utilization, vehicle exhaust and gasoline evaporation, were commonly apportioned as the main contributors of VOCs. In these cities, biogenic sources only made a minor contribution. It should be noted that there are almost no natural sources of VOCs in winter in many cities such as Beijing where coal/ biomass burning was the highest contributor to stamospherioc VOCs (36.6%).[84] However, a unique source category, "aged background air", was found to contribute 24% of the VOCs in Chongqing due to the relatively stagnant air associated with the mountainous nature of the landscape surrounding the city.[86] The different sources determined for these two major cities are further influenced by sampling time, targeted VOC species and meteorological conditions. Detailed quantitative measurements for a consistent suite of VOCs species in aerosols across China, combined with temporal-spatial data, are required to further investigate their sources.

3.4 Particle Number Concentration (PNC)

As a receptor-based model, PMF has not only been widely used to apportion the

sources of particulate chemical composition from filter sampling, but also for resolving

sources of PNC and particle number size distribution (PNSD) data.[88-91] Once particulate

matter is emitted from a given source, its size, number, and chemical composition may

change through multiple mechanisms (for example, by interactions with other chemical

species in the atmosphere) until eventually removed by natural processes.[92]

Additionally, filter-sampling typically lasts for 24 hours or longer, obscuring dynamic

changes in particle size, number and chemical composition, and resulting in a lack of

available emissions information.[93] PNSD and PNC can help to characterize source

contributions and even estimate emission factors.[94] Table 3 summarizes the source

categories of PNC apportioned by PMF in USA, China and UK between 2014 and 2019.

Source contribution for PNC varies among the three countries, but all have a dominant

contribution from traffic source (40%-67%). In the center of Los Angeles, Sowlat et al.,

(2016) showed that traffic sources (type 1 and 2) were the major contributor to PM

number concentrations, collectively making up 60.8 - 68.4% of the total. Their

contribution was also significantly higher in the cold season compared to the warm

season.[95] In New York, although local nucleation and condensation (40 - 46%) and

urban background (31 -36%) contributed most of the PNC, these two categories were

still considered to be associated with traffic emissions.[96] Five periods of measurement

on PNC in Rochester, USA between 2002 and 2016, were compared to assess an

emissions reduction strategy in the northeastern United States. The results showed that

over the 15 years of the study, a general reduction in PNC could be observed. However,

traffic emission overwhelmed (32% - 65%) the other sources, followed by nucleation

(11%   -25%).[97] In Beijing, China, Liu et al., (2014;2015) found traffic and combustion were the two most important sources of PNC on both normal and severe haze days.[98-99] Even during the 2014 Asia-Pacific Economic Cooperation (APEC) summit in Beijing, the contribution from traffic emission was higher (51%) than before the summit (41%).[100] In London, ~40% of PNC were found to be derived from traffic in background areas and at an international airport.[101-102] These results suggested that some changes in the source profiles have occurred since fuel compositions have changed and abatement strategies implemented. In the future, PMF may be extended to other regions, to separate source components and quantify the relative contributions to particle number and mass concentrations in the atmosphere.

3.5 Other contaminants

Source apportionment studies using PMF have also been applied to other contaminants including polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), sulfate (SO_4^{2-}), inorganic nitrogen (NH_4^+, NO_2^-, NO_3^-) and particulate chloride. Taking some examples from 2018, Paromita et al., (2018) investigated polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in surface soils from e-waste recycling sites and near open dumpsites of India. Based on functional activities, PMF identified four sources for PCBs e-waste dismantling, shredding, precious metal recovery and open burning in dumpsites.[103] Qiao et al., (2018) monitored the wet deposition flux of SO_4^{2-}, NH_4^+, NO_2^- and NO_3^- in Jiuzhaigou National Nature Reserve, southwestern China. Four factors

including aged sea-salt, secondary sulfate and nitrate, biomass burning, and dust were resolved using PMF.[104] In China, Qi et al., (2018) focused on NH_4^+ and NO_3^- in atmospheric particles of a coastal city and showed that the contribution of soil dust increased from 23% to 36% on dust days, with decreasing contributions from local anthropogenic inputs and associated secondary aerosols.[105] Also in China, Yang et al., (2018) investigated the sources of particulate chloride and found that coal combustion and residential biomass burning were the main sources (84.8%) of fine chloride in winter, while open biomass burning was the major source (52.7%) in summer.[106]

Our bibliometric analysis showed a multitude of papers on PMF applications to the source apportionment of PAHs, but far fewer with respect to halogenated flame retardants (HFRs) and perfluorinated compounds (PFCs). Contamination of HFRs and PFCs mainly occurred in megacities with large populations and therefore, research on these pollutants was mostly focused on urban areas influenced by intensive anthropogenic activities. Sources of these pollutants were also qualitatively apportioned as mainly coming from industrial production. However, qualitative source apportionment alone cannot cannot provide the data required for control strategies of these pollutants and further quantitative analysis of these compounds is required..

4 Conclusions and Recommendations

Positive matrix fractionation (PMF) has shown thatPAHs in a range of environmental media are mostly derived from petroleum and diesel engine emission, coal burning, wood/biomass burning, and coke oven emissions. Furthermore, the

contribution ratios of these sources are influenced by a number of factors, such as temperature, and transport pathways. Some studies have revealed severe contamination of airborne PAHs in megacities with large populations in China, such as Guangzhou, Shanghai and Beijing, and have also shown significant positive correlations between airborne PAH contamination and detrimental effects on human health, suggesting an urgent need for further study on source control strategies of PAH emissions. Research has also identified high levels of heavy metals as a result of fertilizer applications in agricultural areas and from industrial production. However, heavy metals, are also released to the atmosphere as unintentional by-products of oil- and coal-related production, and these are also important contributors to the heavy metal contamination of the environment.

Our analysis has indicated a knowledge gap for source apportionment of other pollutants, such as PCDD/Fs, PCBs and halogenated flame retardants (HFRs) suggesting the need for PMF applications to these pollutants. In addition to POPs and heavy metals, there is now mounting evidence that exposure to fine particulate matter may lead to increased respiratory and cardiovascular diseases, cardiovascular morbidity and mortality.[107] Most studies have focused on using the PMF model to determine the sources of chemically specified PM mass, but recently, it has also been applied to PNSD data to characterize sources of increasing concern.[108] As complex chemical composition of particulates may obscure a deeper understanding of its source apportionment, investigating high-resolution particle number size distribution (PNSD) and particle chemical composition (PCC) data in separate or combined PMF runs may help identify

and apportion contribution. However, selection of relevant parameters, combinations of PMF runs and comparison and analysis of PMF results are different between many current studies. These may impact accuracy and precision or even cause error in source apportionment of PNC. It is important to highlight that we should further complete and develop new metrics, such as PNC, in atmospheric pollution criteria. Then, standardized results could be used in future epidemiological studies to link particulate matter sources to adverse health effects as well as by policymakers to set targeted and more protective emission standards. In addition, different source apportionment models may generate different outcomes as a result of different algorithms within the models and estimations of uncertainty. Applying multiple techniques that include, but are not limited to PMF, to determine sources can minimize individual-method weaknesses and thereby increase confidence in results.

In general, coal combustion and vehicular emissions appear, from most source apportionment results, to be the major sources, of many pollutants. Fossil-fuels are likely to remain the main global energy source over the next few decades. In 2015, fossil-fuels accounted for 81% of the global energy structure and, although it is expected that this proportion will decline, by 2040 it is still expected to be 74% (https://www.opec.org/opec_web/en/publications/338.htm). The continued dependency on fossil-fuels will remain an important cause of environmental pollution problems. Developing renewable sources such as wind, geothermal and solar power will improve the energy structure and establish a sustainable energy system to both strengthen environmental protection and promote economic development.[109]

Acknowledgements

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Table captions

Table 1 Source categories of heavy metals in soil and road dust in China in 2018 as apportioned by PMF.

Table 2 Source categories of VOCs in the atmosphere in 2018 as apportioned by PMF

Table 3 Source categories of PNC in USA, China and UK between 2015 and 2019 as apportioned by PMF

Figure captions

Figure 1 The frequency of PMF model use in recent years for PAHs, heavy metals, anthropogenic aerosols and VOCs.

Figure 2 PMF application status concept map.

Figure 3 The ratio of source apportionment of PAHs, heavy metals and VOCs from 2002 to 2018 both globally   and in China using PMF. (*China is divided into eight regions,*

including East China (EC): Jiangsu, Zhejiang, Anhui, Fujian, Shanghai, Shandong; South China (SC): Guangdong, Guangxi, Hainan; Central China (CC): Hubei, Hunan, Henan, Jiangxi; North China (NC): Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia; Northwest China (NWC): Ningxia, Qinghai, Xinjiang, Shaanxi, Gansu; Southwest China (SWC): Sichuan, Yunnan, Guizhou, Chongqing, Tibet; Northeast China (NEC): Liaoning, Jilin, Heilongjiang; Hong Kong, Macau and Taiwan (HMT).

Figure 4 The frequency and distribution of PMF model use in recent years for Particle Number Concentrations.

Figure 5 The source categories of PAHs and their contributions in the solid phase in Asia as apportioned by PMF.

Figure 6 The source categories of PAHs in the air (both particulate and gas phase), apportioned by PMF.