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REVIEW

Mapping sources of atmospheric pollution: integrating spatial and cluster bibliometrics

Shudi Zuo, Panfeng Dou, and Yin Ren

Abstract: The intensified degradation of regional air quality in recent years has prompted an increase in research teams to pursue various aspects of air pollution research including source apportionment (SA) resulting in rapidly growing output of this literature, a rapid shift in research emphasis, and an extensive inter-institutional and international collaboration. To explore these recent trends in air pollution research, we present a spatial and cluster bibliometric evaluation of atmospheric SA research, ascertaining the relationship among the main research bodies and knowledge clusters. The evolution trend of the knowledge clusters was depicted with an emphasis on the recent surge of research in Asia. We retrieved relevant articles from the Web of Science and Scopus data set with the key words "air" and "source apportionment". In this field, the annual output of peer-reviewed papers has increased dramatically since 2005. Initially, air SA research was concentrated in developed countries from Europe and North America, but more recently it has been receiving substantial contributions from developing countries, most notably China and India. Meanwhile, there has been a methodological shift from single methods, such as source diffusion models or receptor models, to the integration of online high-resolution data monitoring with multiple models. In recent years, the main research objectives for atmospheric pollution research have changed from being focused predominantly on coarse particles to more focus on fine and ultrafine particles. Our bibliometric analysis yielded 666 knowledge nodes, forming 26 major co-citation clusters. Focusing on recently emerged knowledge clusters 7, 22, and 23, we identified four research themes based on the research frontier analysis: (*i*) practical research conducted to inform pollution-reducing policy, (*ii*) assessment of pollution-related health risks of human activities, (*iii*) analysis of pollutant composition and formation, and (*iv*) improvement of SA methods. The study provides an efficient and economical means to understand the history and the prospects for air SA research through a temporal and spatial analysis on the relationships among the main research bodies and knowledge clusters.

Key words: receptor modeling, research trends, air pollution, particulate matter, aerosol, Asia.

Résumé : L'intensification de la dégradation de la qualité de l'air à l'échelle régionale au cours des dernières années a incité un plus grand nombre d'équipes de recherche à approfondir divers aspects de la recherche sur la pollution atmosphérique, y compris la répartition des sources (RS) occasionnant la croissance rapide de production de cette littérature, un changement rapide des priorités de la recherche et une collaboration interinstitutionnelle et internationale importante. Afin d'explorer ces tendances récentes en matière de recherche sur la pollution atmosphérique, nous présentons une évaluation bibliométrique spatiale et par grappes de la recherche sur la RS de pollution atmosphérique, ce qui permet de déterminer la relation entre les principaux organismes de recherche et les grappes de connaissances. L'évolution de la tendance des grappes de connaissances a été décrite, en mettant l'accent sur la récente vague de recherche en Asie. Nous avons extrait des articles pertinents de l'ensemble de données « Web of Science » et « Scopus » avec les mots-clés « air » et « répartition des sources ». Dans ce domaine, la production annuelle de documents évalués par les pairs a augmenté de façon phénoménale depuis 2005. Initialement, la recherche sur la RS était concentrée dans les pays développés d'Europe et d'Amérique du Nord, mais elle a récemment reçu des contributions substantielles de pays en développement, notamment de la Chine et de l'Inde. Entre-temps, il y a eu un virage méthodologique qui est passé de méthodes uniques, comme les modèles de diffusion à la source ou les modèles de récepteurs, à l'intégration de la surveillance en ligne de données à haute résolution avec de multiples modèles. Au cours des dernières années, la portée des principaux objectifs de la recherche sur la pollution atmosphérique a évolué d'une concentration sur les particules grossières à une concentration sur les particules fines et ultrafines. Notre analyse bibliométrique a produit 666 nœuds de connaissances, formant 26 grandes grappes de co-citations. En nous concentrant sur les grappes de connaissances 7, 22 et 23 récemment créées, nous avons identifié quatre thèmes de recherche fondés sur l'analyse de la recherche exploratoire: (*i*) la recherche pratique menée pour éclairer la politique de réduction de la pollution; (*ii*) l'évaluation des risques sanitaires de la pollution lié aux activités humaines; (*iii*) l'analyse de la composition et de la formation des polluants et (*iv*) l'amélioration des méthodes de RS. L'étude procure un moyen efficace et économique de comprendre l'historique et les perspectives de la recherche sur la RS dans l'air par une analyse temporelle et spatiale des relations entre les principaux organismes de recherche et les grappes de connaissances. [Traduit par la Rédaction]

Mots-clés : modélisation des récepteurs, tendances de la recherche, pollution atmosphérique, particules, aérosols, Asie.

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1. Introduction

Growing evidence indicates that exposure to high concentrations of air pollutants results in increased risks of a number of potentially lethal chronic diseases including cardiac and respiratory diseases (Lanki et al. 2006; Sarnat et al. 2008; Wei et al. 2010; Masiol et al. 2012; Singh and Gupta 2016). Air pollutants include a variety of constituents, such as heavy metals, particulate matter (PM), gaseous pollutants (e.g., sulfur dioxide, ozone, nitric oxide, and nitric dioxide, etc.), and persistent organic pollutants (e.g., dioxins, endosulfans, polychlorinated dibenzofurans, polychlorinated biphenyls, etc.). Pollution prevention and control programs are informed by pollutant source analyses. Source apportionment (SA), wherein pollution sources and environmental impacts are determined, has been employed in atmosphere, water, and soil pollutant source identification (Wang et al. 2012). Thus far, atmospheric SA models have focused mainly on PM source analysis based on data produced by atmospheric monitoring or fixed-point sampling (Hopke 2016). Employing these approaches, researchers can delineate the contributions of major pollutant sources to extant pollution contents. Thus, SA can be used to develop targeted prevention initiatives, well-reasoned policies, and reasonable regulations.

SA methods, which have been evolving over the last half century, can be classified into three categories: source emission inventory methods, diffusion modeling of pollutant sources, and receptor modeling of pollution sites (Viana et al. 2008). SA technology encompasses a complex system of methods with differing strengths, applicability, uncertainty levels, and limitations (for details on the advantages and limitations of the three categories, see Supplementary Material[1]). Scientific reviews are normally compiled by experts who vigorously summarize the component proportions of atmospheric pollutants, possible pollution sources, and pollution sites based on their professional knowledge, reading, and and research (Belis et al. 2013; Banerjee et al. 2015; Chow et al. 2015). Many popular reviews emphasize reporting on a detailed description on the origins, developments, and technical methods of future research directions, especially for hot research topics. (de Gennaro et al. 2014; Vu et al. 2015; Vorkamp 2016). However, since annual publication rates for SA research increased substantially between 2005 (<100 articles) and 2016 (>400 articles), with much of the research sites having shifted to Asia, this field of study is now considered to have entered the phase of rapid development. During this period, the research emphasis transferred faster than before. Furthermore, the influence of air pollutants on regional scales directly involving extensive stakeholders have led to a greater the inter-institute and international cooperation. Recent review articles on SA research commonly focus on popular technologies for a pollutant, and a more comprehensive review is rare given that it is more labor-intensive.

A bibliometric evaluation of the many studies of SA techniques could serve as an alternative and innovative way of understanding the techniques' applications, which may potentially reveal global trends in atmospheric pollution research (Xu and Boeing 2013). This method has been used in a variety of study fields, such as drinking water research and urban heat island effects (Fu et al. 2013; Blank et al. 2013). However, a change in the number of citations, key words, or publications cannot provide a complete summary of development trends for a research field (Chiu and Ho 2007). The drivers behind the change are more attractive for the readers. Therefore, the title, key words, abstracts, and co-citation network (Chen 2017) should be introduced in the study of the research trend.

In this paper, we aimed to provide readers with perceptual knowledge of the constitutional structure in atmospheric pollution SA between 1980 and 2016 using statistical algorithms, spatial visual software, and cluster analysis. Specifically, we first provide an analysis of annual SA manuscript output over time and characterize research stages based on numbers of publications, authors, pages, citations, key words, and references. We then present a global distribution map of the top 30 countries and institutions in terms of publication volume, an analysis of the cooperation frequencies among them, and a detailed analysis of the sources of the greatest number and highest quality publications. Third, we rank the most cited journals, indicating the main research aims, methods, and focuses based on key words and citation index values. Finally, we present our findings with respect to the highest frequency key words and their correlations. We conducted cluster analysis in Citespace software to show the research focus has evolved, including the identification of recent research themes. We include an analysis of recent SA studies specific to Asia and the research interests that these studies suggest.

2. Bibliometric data and analysis

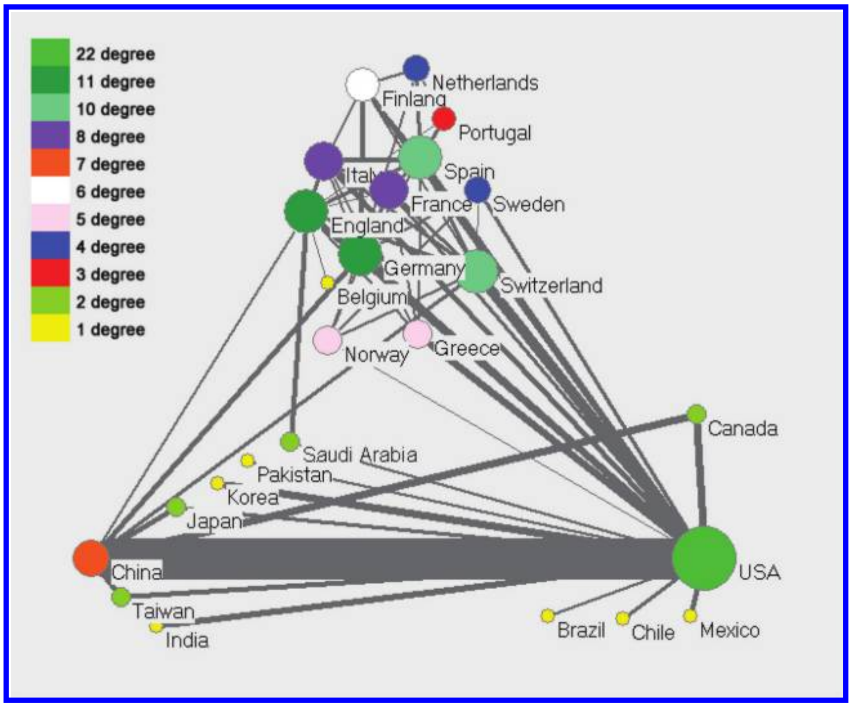
We used bibliometric publication statistics for air-SA technology to produce a network diagram. We conducted a search for the key words "air" + "source apportionment" to retrieve publications of interest appearing between 1980 and 2016 on the Web of Science and Scopus databases. The records of 4765 articles were retrieved, including 40 descriptors such as: authors, title information, language, key words, author address, publication year, corresponding authors, number of references, number of citations, page count, subject category, and journal name, among others. The citation rate data reported here were updated on 1 November 2016.

We followed the method of article classification described by Xu and Boeing (2013). Briefly, "single article" classifications refer to studies by researchers who were all from the same country or institution; "international article" refers to studies by researchers from at least two countries or institutions (Xu and Boeing 2013). We explored development trends using key word frequencies and highly referenced articles, and we used Citespace software to identify knowledge clusters.

Cooperation activities were mapped in Pajek 4.09 based on a co-occurrence analysis of the top 30 published and cited countries, institutions, and key words in BibExcel (Persson et al. 2009). The net files generated via pairing of items in the software's list box reveal inter-item correlations, which can be used in further analysis and visualization in Pajek. Citespace integrates cluster, social network, and multi-dimensional scale analysis methods to monitor and analyze trends at the forefront of research, the relationship between this research and its knowledge bases, and relations among frontier research fields. Each research field is conceptualized as a time map from its frontier to its knowledge base, i.e., the original research reports in the literature (Nieminen et al. 2013). Therefore, the clusters and evolution of the knowledge base form the basis for analyzing research frontiers; such analyses can reveal important turning points in research and clarify how frontiers inter-relate across fields (Jensen et al. 2016). In this study, the knowledge structure atlas for the air-SA field was based on the data in the publication citation network. The atlas can be used to analyze the theoretical structure of the field of air-SA. The parameters employed by Citespace were: time-cutting period setting, "2*a*"; theme source; key words; title; summary; line word; identifier; theme type; "noun phrase"; node type, "cited document"; and threshold, top 50.

[1]Supplementary data are available with the article through the journal Web site at http://nrcresearchpress.com/doi/suppl/10.1139/er-2018-0105.

Fig. 1. Collaboration networks for countries with over 10 research partners. The color indicates the primary partner number of the country. The partners with less than five cooperative articles do not display. The width of the lines represents the frequency of cooperation between two countries.



3. Profile of publication activity

3.1. Publication output and journal popularity

Most of the 4765 articles (n = 4718; 99.03%) were published in English. During the study target period (1980–2016), the annual number of articles in the field of air-SA increased from 1 per year in the early 1980s to 809 in 2016. This trend became evident in the early 1990s and accelerated after 2005 (see Supplementary Fig. S1[1]). Based on the annual numbers of articles and key word frequency cluster analysis, we divided the development of air-SA research into four stages: pre-1990, 1991–2005, 2006–2013, and 2014 onward.

The top 20 journals, accounting for 90% of the articles, are indicated in Supplementary Table S1[1]. *Atmospheric Environment* published most articles, followed by *Atmospheric Chemistry and Physics* and *Environmental Science & Technology*. Other highly cited journals in the field (citation rate > 30) include *Environmental Health Perspectives* and *Chemosphere*.

Approximately half the number of top 20 articles appeared in the top three journals, indicating a narrow focus of interest in the air-SA technique. We did not consider self-citation in our analysis, because while progress is grounded in previous achievement, it is difficult to identify meaningful self-citation in this context. Additionally, our analysis of the co-citation network of articles below incorporates self-citation.

The most frequently cited article in *Atmospheric Environment* described chemical mass balance (CMB) receptor modeling and the main research topics addressed in very highly cited articles are related to particulate matter (PM), aerosols, $PM_{2.5}$ (≤2.5 μm), polycyclic aromatic hydrocarbons (PAHs), PM_{10} (≤10 μm), chemical composition, volatile organic compounds (VOCs), and the positive matrix factorization (PMF) method. The common key terms shared among the most popular articles in *Atmospheric Chemistry and Physics* include PM, high resolution, chemical composition, aerosol, and PMF. The most cited articles from this journal focused on seasonal analysis of $PM_{2.5}$ SA and its chemical characteristics in Beijing using CMB, PMF, trajectory clustering, and potential source contribution functions to characterize aerosol speciation. Common key terms in *Environmental Science & Technology* are PMF, PM, PAH, aerosol, organic compounds, and $PM_{2.5}$. Based on principal component analysis (PCA) with multi-linear regression analysis, the most highly cited article indicated that a combination of PAH and mechanical pollutant measures provided a more powerful tracer of emission sources than PAH data alone.

3.2. Countries and collaborations

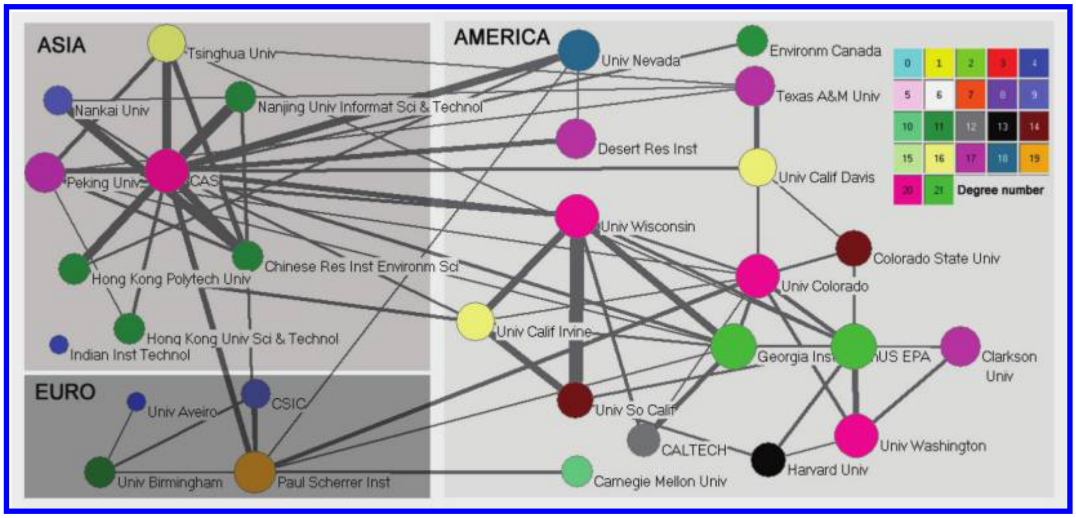
A total of 114 countries contributed to the air-SA research literature during the study period, with the largest number of articles originating in the United States (3306/articles, 28.95%), followed by China (2206, 19.32%), Italy (500, 4.38%), Spain (473, 4.14%), and Germany (331, 2.90%). Aside from the five highest output countries, India, England, and Switzerland also have average h-index values (mean Hirsch numbers) of over 30 (Supplementary Table S2[1]). These data indicate that these countries produce air-SA research high in both quantity and quality.

Countries with more than 10 internationally collaborative articles are shown in Fig. 1, which shows substantial collaboration among researchers in Asia, America, and Europe. Researchers from the United States published with counterparts from 28 countries, with US–China collaborations prominent (n = 217). The United States and China have emerged as current collaborative centers in the Americas and Asia, respectively. Interestingly, while Spain (26 collaborating countries) and China (24 collaborating countries) collaborated widely, they rarely did so with each other (n = 3). Cooperation among European countries is evident, led by Spain, followed by Switzerland, England, and Germany.

3.3. Institutions and collaborations

The top 30 research institutions with respect to publication are indicated in Supplementary Table S3[1]; the top three among them are the Chinese Academy of Sciences (CAS), which has 114 institutes or research centers (456 articles); US Environmental Protection Agency (EPA), which has 74 offices, labs, and research centers

Fig. 2. Collaboration networks of the 30 most productive institutes. The color indicates the number of research partners for each institute. The width of the lines represents the frequency of cooperation between countries.



(177 articles); and the University of Wisconsin with 13 schools and colleges (161 articles). These top 30 institutions all prefer inter-institution collaborations as opposed to single-institution articles. Half of the institutions have more average citations per article for their inter-institution collaborative articles than for their single institution articles, including the EPA, Georgia Institute of Technology, and Peking University. The top six institutions in terms of publication have *h*-index values greater than 30. Additionally, the University of Colorado (*h*-index = 38), Carnegie Mellon University (*h*-index = 36), University of Washington (*h*-index = 35), and CALTECH (*h*-index = 31) contributed studies to the literature that are noteworthy for their high-quality ratings.

As can be seen in Fig. 2, US EPA and Georgia Institute of Technology have the highest number of collaborators, each having 21 partner organizations. Following CAS, the Universities of Wisconsin, Colorado, and Washington have more than 20 partners each. The Institute of Earth Environment, Guangzhou Institute of Geochemistry, Institute of Atmosphere Physics, and the Eco-environmental Sciences Research Center are the major organizations in CAS, which collaborates mostly with Chinese institutions; its most common foreign partner is the Desert Research Institute of the University of Nevada. Similarly, the University of Wisconsin collaborates mostly with US institutions and occasionally with CAS. In Asia, a research cluster centered on CAS, Nanjing University of Science & Technology, and the Chinese Resource Institute of Environmental Science has formed. In North America, there is a research node centered on the University of Wisconsin, the EPA, and the University of Southern California. Within Europe, the Paul Scherrer Institute in Switzerland has emerged as a focus of inter-institutional collaboration. Overall, these collaborations reflect the common research interests and mutual understandings of research teams.

4. Evolution paths based on key word and knowledge cluster networks

4.1. Key word analysis

Changes in key word frequencies over the three phrases of the study reveal changing research interests. Citespace's word cluster algorithm was used to identify the evolution paths of knowledge clusters. The algorithm places the term "source apportionment" at the center of the key word network analysis. The remaining key words are divided into four categories: objects analyzed (i.e., type of pollution), object characteristics, methods, and research sites (Fig. 3). Most of the key words are research objects, such as PM, $PM_{2.5}$, aerosol, PAHs, PM_{10}, and dust. The terms $PM_{2.5}$, PM_{10}, organic carbon, coal, elemental carbon, and especially PM_1 show dramatic increases. The terms particulate matter, aerosol, dust, exhaust gas, and volatile organic compound(s) received attention over the whole period. The term biomass burning emissions is prominent, and the method term environmental monitoring has increased in rank from 19–23.

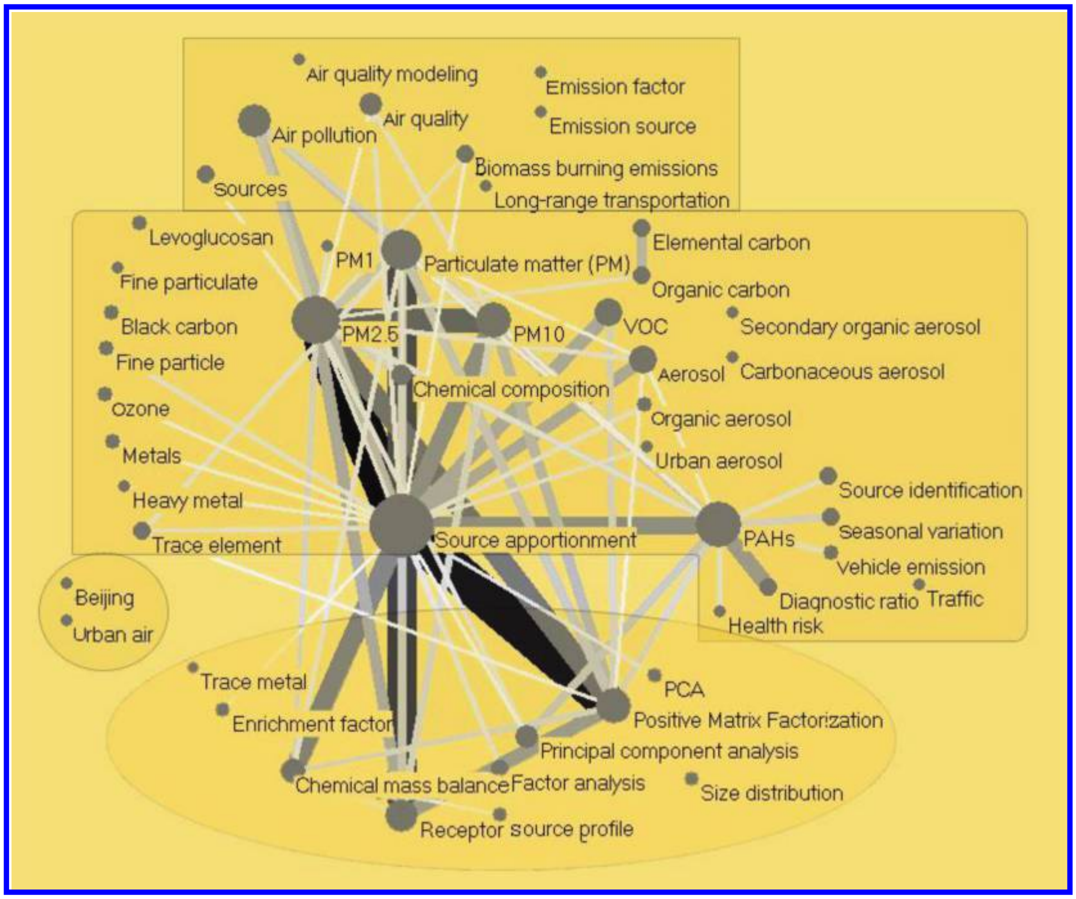
4.2. Knowledge cluster analysis

Bibliometric meta-analyses can reveal geographical distributions and relations of data and their concentration around specific clusters or points (Chen 2006). Combining the centrality concept of graph theory with co-citation network analysis, which involves automated text summarization and natural language processing algorithms, may reveal research interests and rapidly highlight new scientific trends (Chen et al. 2012). Our Citespace cluster analysis yielded 666 nodes inter-connected by 1188 lines, forming 26 major co-citation clusters. The modularity of the network is 0.91, indicating accurate network clustering. The clusters are numbered from the most to fewest articles in the form cluster 0, 1, 2, etc. The major 26 clusters are shown in Fig. 4 (see Supplementary Table S4[1] for all 26 clusters).

Barone et al. (1978) pioneered the receptor model in a study demonstrating the importance of particle size in determining the effects of aerosols on visibility reduction (Barone et al. 1978). In the 1980s, there were clusters 3, 5, 6, 11, 17, 19, and 21. The highest cited study during this period had used physical methods and employed cluster analysis encompassing a variety of data set aspects, such as data processing, cluster algorithm optimization, error analysis, cluster composition, and uncertainty to analyze individual particles and the sources (Van Borm and Adams 1988).

The nodes in cluster 0 show high centrality and are closely related to the surrounding literature, with concentrated connection lines forming a cohesive research network. During this period, there was surge in research on black carbon, organic carbon (OC), elemental carbon (EC), and ions in air pollution. The early work in this cluster was conducted primarily in major cities in the Americas and Europe, and later work came from China (e.g., Beijing, Tianjin, Shenzhen, and Guangzhou), India (e.g., Delhi), South Korea (e.g., Seoul), Taiwan (e.g., Taichung), and Iran (e.g., Tehran). Most articles in this cluster were based on a single physical method of analysis, predominantly gas chromatograph – mass spectrometer (GC/MS), X-ray fluorescence, or CMB; a small number of studies used ion chromatography (IC), PMF, and PCA, whereas others employed a combined PMF-CMB methodology (Zheng et al. 2006; Bullock et al. 2008). The most cited articles in

Fig. 3. The co-words network of the 50 most frequent keywords. The size of the circle represents the frequency of appearance of the keyword. The width and gradient color of the lines represent the degree of connection between word pairs: the wider and darker the line, the closer the connection. The frame and the circle contain the same types of information associated with the word, such as the characteristics of the pollutants, pollutant element, method, and site. The keywords fine particle and fine particulate were merged in PM2.5.



the cluster are on the use of organic tracers to identify sources of airborne PM and secondary organic aerosol (SOA); these articles advanced methodology and thinking in the field.

Clusters 2, 7, 22, and 23 appear to have formed as serial offshoots of clusters 0 and 8, each becoming highly cited albeit through inter-linking citation networks. The age of cluster 2 citations, and its high citation rates compared to cluster 7, affirm our inference that cluster 0 gave rise to cluster 2, which in turn gave rise to clusters 7, 22, and 23.

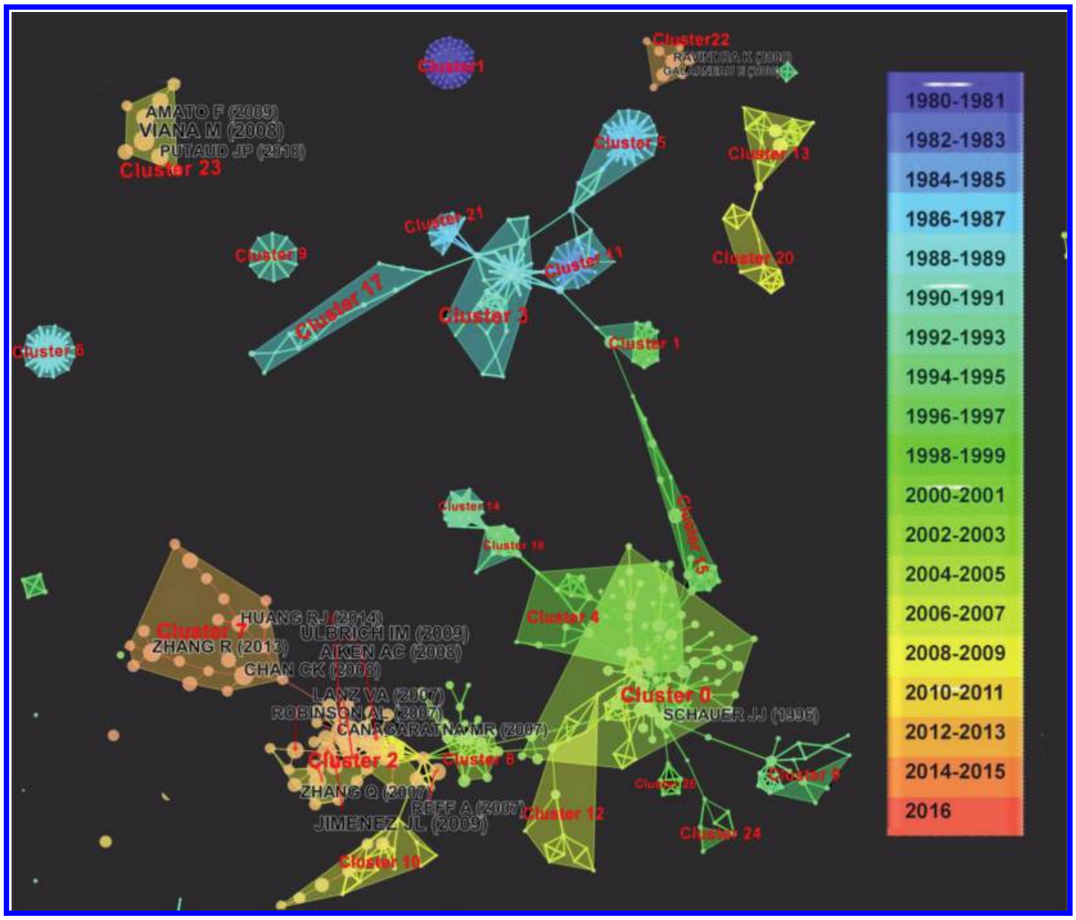
We have recently observed approaches using source and receptor modeling to overcome the limitations of using either one alone, and the combining and use of different methods appears to be a feature of PM analysis. Since these clusters (2, 7, 22, and 23) will probably ground emerging conceptual frameworks and theory in this field, we discuss them in further detail below.

4.2.1. Towards combined methods—cluster 2

Key articles in cluster 2 center on analysis of the chemical composition of aerosol/organic aerosol (OA) pollutants (especially from traffic vs. industrial sources), evolutionary processes, and harm to human health, with research mainly from Italy, France, Spain, and Greece within Europe, and in the United States, Canada, and Mexico in the Americas (Canagaratna et al. 2007; Lanz et al. 2007; Reff et al. 2007; Robinson et al. 2007; Aiken et al. 2008; Jimenez et al. 2009; Ulbrich et al. 2009). Urban, regional, and global scales are included. The research topics of cluster 2 are aerosol pollutants and the challenges associated with air-SA technology, especially with respect to distinguishing between anthropogenic and natural pollutant sources. More than half of the cluster 2 articles report aerosol data by way of combined methods. Specifically, for source tracing, researchers combined aerodyne time-of-flight aerosol mass spectrometry (ToF-AMS) with proton transfer reaction-MS (Slowik et al. 2010), inductively coupled plasma mass spectrometry (ICP-MS) with GC/MS (Cheung et al. 2010), and aerosol MS techniques (i.e., quadrupole AMS, ToF-AMS) with nuclear magnetic resonance spectroscopy (Finessi et al. 2012). In the calculation phase, researchers combined back trajectory analysis with PMF (Freney et al. 2014), PMF with multi-linear methods, PMF with FAs (van Drooge et al. 2012; Venturini et al. 2014; Zhang et al. 2014), PMF with CMB (Zhang and Ying 2010), CMB with FAs (Sun et al. 2012; Poulain et al. 2014), and even PMF with both FAs and PCA (Bialek et al. 2014; Presto et al. 2014). Overall, the most common research methods employed in cluster 2 studies were PMF, CMB, factor analysis (FA), and high-resolution ToF-AMS.

Innovative methods also characterize Cluster 2. The authors critically evaluated existing methods, with many deriving their own. For example, Finessi et al. (2012) used nuclear magnetic resonance spectroscopy for organic functional group characterization of the polar organic fractions of aerosols. Using high-resolution ToF-AMS data with VOC tracers from a proton transfer reaction-MS method to estimate the relative importance of secondary versus primary OAs (SOAs vs. POAs), Liggio et al. (2010) showed that SOAs account for a greater proportion of total OA pollution than POAs. Indeed, high-resolution ToF-AMS (HR-ToF-AMS) has also been used in combination with PMF to analyze POAs, SOAs, and inorganic species in aerosols, providing insights

Fig. 4. Major clusters in the co-citation analysis of references. The different colors represent the date of publication. Node sizes represent the citation frequency of articles. The line represents co-citation. Articles with more than 70 citations are marked with author and publication date.



into the sources, transfer processes, and chemical characteristics of aerosols in the atmosphere (DeCarlo et al. 2006; Sun et al. 2012).

Receptor modeling has become a mainstream method of analysis in the SA calculation phase. For instance, PMF with multilinear engine (version 2) software improves the efficiency of identifying various, predominant OA sources (Canonaco et al. 2013; Crippa et al. 2014). PMF of AMS organic mass spectra (Paatero and Tapper 1994; Lanz et al. 2007; Ulbrich et al. 2009) can be used to differentiate between OA sources and formation processes. However, PMF cannot distinguish between anthropogenic and biogenic OAs, nor between fossil fuel derived and nonfossil fuel derived OAs, especially in analyses of SOAs. Zotter et al. (2014) used radiocarbon (^{14}C) analysis to distinguish fossil/nonfossil fuel sources of carbonaceous aerosols. Daellenbach et al. (2016) proposed an offline technique for real-time AMS analysis, which may enhance AMS applicability to size-fractionated, spatially resolved, long-term datasets.

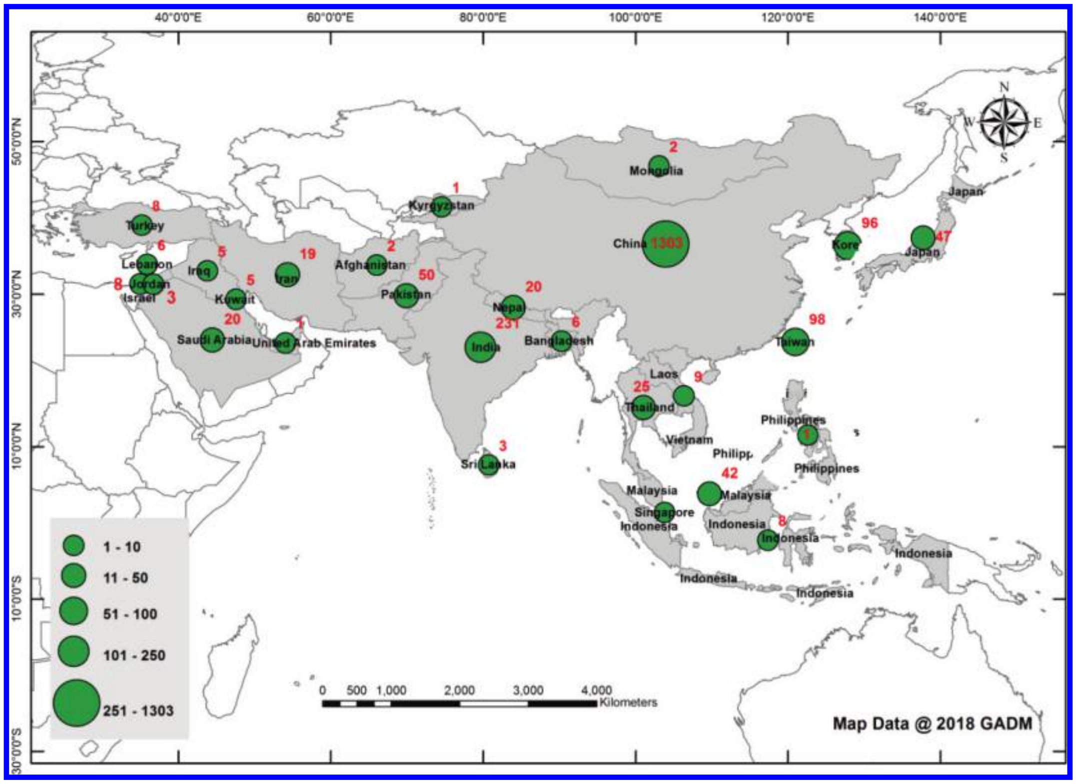
Diffusion modeling has also improved in recent years. Several new techniques have been developed to better enable large-scale data analysis of targeted compounds, such as the detailed three-dimensional regional/global chemical transport model PMCAMx (comprehensive air-quality model with extensions, modified for application to PM) (Fahey and Pandis 2003; Gaydos et al. 2003), MOZART-4 (model for ozone and related chemical tracers, version 4), and the EOS-Chem (Aura satellite data) model. Researchers have used the PMCAMx model to calculate local percentages of $PM_{2.5}$, EC, POA, SOA, SO_2, and VOC pollutants, comparing their results with those obtained from PMF or AMS-PMF (Tsimpidi et al. 2010; Skyllakou et al. 2014; Fountoukis et al. 2014). Emmons et al. (2010) used MOZART-4 to obtain a high-resolution (70 km) model of the air quality in Mexico City, and validated their results with empirical data from the MILAGRO (Mega-city Initiative: Local and Global Research Observations) project. Pye and Seinfeld (2010) used the EOS-Chem model to estimate levels of VOC subtypes (semi-volatile and intermediate-volatility organic compounds) in the atmosphere and to analyze their sources on a global scale.

4.2.2. Prospects—clusters 7, 22, and 23

The research topics in clusters 7, 22, and 23 are more varied than those in cluster 2, and include particulate matters (Clusters 7 and 23), black carbon (Cluster 22), PAHs (Cluster 22), aerosols (Cluster 23), and road dust (Cluster 23), as well as SA technology (Chan and Yao 2008; Ravindra et al. 2008; Zhang et al. 2013). The research sites are concentrated in the most polluted regions of Asia, including cities in China, Taiwan, and India. Applying the "burst detection" in Citespace, the following four research direction themes (I-IV) emerged, based on our reading of the articles in clusters 7, 22, and 23:

(1) *Practical research to inform pollution-reduction policy*. These studies used SA technology to analyze pollutant characteristics, targeting critical urban pollution. The inventory of air pollutants emission serves as a foundation based upon which efficient mitigation policies are created (Guo et al. 2014; Zhang et al. 2009). Recommendations from this research included: use of dust control technology to reduce metal concentrations in urban air (Tan et al. 2014), regional policies to address the transmission characteristics of regional pollution (Zhang et al. 2015), unification of wind patterns and detailed the size-resolution information of other source data (Tian et al. 2016), empirical assessment of the efficacy of pollution control methods (Ahmed et al. 2016), and

Fig. 5. Study sites and their reported frequencies in Asia. The red numbers represent the reported frequency.



enforcement of regional carbon aerosol and VOC emission regulations to achieve Beijing's $PM_{2.5}$ and ozone reduction goals (Ji et al. 2016; Song et al. 2007).

(2) *Assessment of pollution-related health risks*. Health-risk analyses related to air pollution are currently limited, especially in developing countries with concentrated and complex pollutant sources (Liu et al. 2014). Researchers noted an urgent need to standardize risk assessment parameters for given activities and to improve the reliability of risk assessments (Izhar et al. 2016). It is hoped that assessments of PM levels at specific sources and of the health risks posed by individual hazardous elements will improve our understanding of exposure routes and the risk factors of carcinogenic and noncarcinogenic substances (Khan et al. 2016). Further development of biomarkers is expected to improve health risk assessments for $PM_{2.5}$ and PM_1 exposure (Pant et al. 2016). Studies are also needed to clarify the contributions of social (e.g., gender, socioeconomic status) and lifestyle (e.g., relative time spent in different microenvironments) factors as predictors of risk (Pant et al. 2016).

(3) *Analysis of pollutant composition and formation*. This is a long-studied field. Nevertheless, there are complex aspects of pollutant composition and formation for which information is lacking, most notably the formation of organic matter pollutants and the chemistry of secondary pollutant formation, including changes in toxicity that accompany the conversion of primary pollutants into secondary pollutants (Pant et al. 2016). Researchers have called for investigation of the factors driving the transformation of iron species and their impacts on biochemistry (Chan et al. 2016). Furthermore, it is important to consider the interactions between meteorological parameters (e.g., seasonal data, radiative forcing etc.) and air pollutants (black carbon, PM etc.) (Bond et al. 2013; Hooda et al. 2016; Ramanathan et al. 2008).

(4) *Improvement of SA methods.* Analytical and statistical approaches to investigating the chemical nature and sources of pollutants hold promise for improvement e.g., CMB-PMF with ME-2 (Huang et al. 2014; Viana et al. 2008). Gao et al. (2016) suggested that including measurements of silicon and aluminum species would improve the stability of receptor models with online data sets. Uncertainty in the model predictions could be reduced through accurate descriptions of spatial variation in PM concentrations and pseudo Monte Carlo simulations, better methods to estimate secondary products, and the development of ensemble SA models (Lv et al. 2016; Huang et al. 2014). Nondestructive methods of PM analysis would improve the resolution of PM source tracing (Ji et al. 2015). Moreover, the identification of organic compounds that can be used as tracer substances for sources of pollution will help to clarify the proportions of various source emissions (e.g., gasoline vs. diesel vs. biomass vs. waste incineration) (Kim et al. 2015).

5. Findings from Asian cities

Of the 4765 articles retrieved, 1759 were studies from Asia. The publication data indicate that all but 39 of these 1759 studies were published after 2000. Chinese cities are mentioned 1318 times (including mentions of multiple cities within single articles). After China, the second most studied region is India (230 papers). Overall, the research sites were concentrated in eastern Asia including estuarine Delta urban agglomerations, India, Taiwan, South Korea, Japan, Pakistan, Malaysia, and Nepal. The relatively few studies from western Asia were mostly from Saudi Arabia, Jordan, Israel, and Kuwait. Various geographical features received attention, including straits, mountains, plateaus, lakes, and nature reserves (Fig. 5).

Focusing on clusters 7, 22, and 23 in the research frontiers of air-SA in Asia, and Beijing in particular (43 articles), we found that, consistent with our key word frequency analysis, $PM_{2.5}$ (38 articles), PM_{10} (10 articles), and PM_1 (6 articles) were the most important target metrics of the studies in Beijing (Fig. 6*a*). PM_1 could become the next pressing research topic. PM_{10} (19 articles) and $PM_{2.5}$ (18 articles) were also commonly studied in Asian sites outside of China (Fig. 6*c*). The most frequently used method among air-SA studies conducted in Beijing was PMF, followed by IC, ICP-MS,

Fig. 6. (*a*) Target metric of SA studies reported in Beijing (Clusters 7, 22, and 23). (*b*) Methods used in SA studies reported in Beijing (Clusters 7, 22 and 23). (*c*) Target metric of SA studies reported in other Asian cities excluding Beijing (Clusters 7, 22 and 23). (*d*) Methods used in SA studies reported in other Asian cities excluding Beijing (Clusters 7, 22 and 23). Legends were composed of the name and the proportion of each section to the all 43 Beijing articles (Supplementary Table S5[1]).

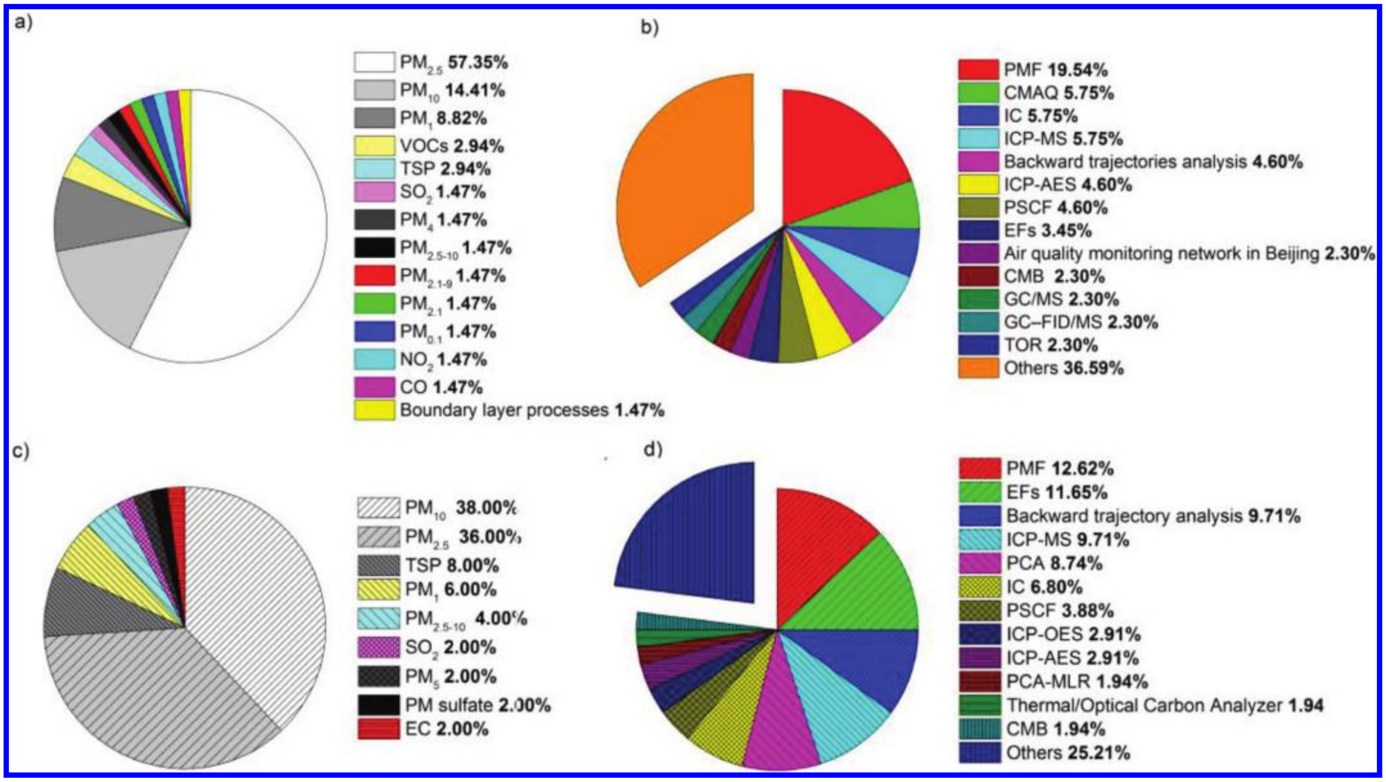
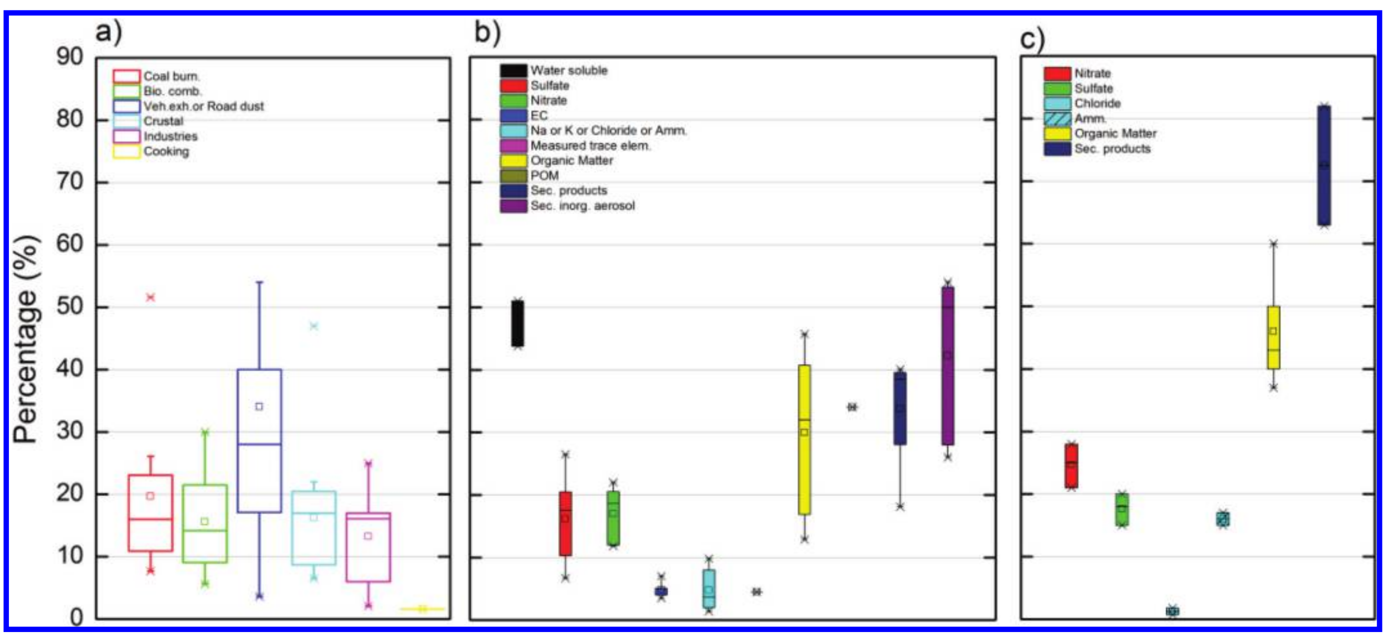


Fig. 7. Median, interquartile range (boxes), and minimum and maximum (whiskers) relative contributions from each source category to: (*a*) $PM_{2.5}$ sources in Beijing, and (*b*) $PM_{2.5}$ composition in Beijing, and (*c*) PM_1 composition in Beijing. Coal burn., coal burning; bio. comb., biomass burning; veh. exh., vehicle exhaust; crustal, re-suspended dust; amm., ammonia; sec., secondary; Inorg., inorganic.



community multi-scale air quality models, backward trajectory analysis, ICP-atomic emission spectrometry, potential source contribution functions, and enrichment factor analysis (EF) (Fig. 6*b*). Studies conducted at Asian research sites outside of Beijing commonly employed EFs and PCA (Fig. 6*d*).

For the results of studies in Beijing during the study period of cluster 7, 22, and 23, we created a picture of pollutant components and dominant pollution sources in Beijing (Fig. 7; data source from the articles of Supplementary Table S5[1]). The greatest source of $PM_{2.5}$ in Beijing (Fig. 7*a*) is traffic (vehicle exhaust and road dust, 34.64 ± 5.41%, mean ± SE), followed by coal combustion (19.67 ± 4.49%), crustal dust (16.27 ± 2.65%), biomass combustion (15.64 ± 3.00%), and industry (13.24 ± 4.11%); the least significant source indicated by these studies is cooking (single report, 1.60%). Regarding the composition of pollutants in Beijing (Fig. 7*b*), the reports indicate that $PM_{2.5}$ consists mainly of secondary inorganic ions (mean ± SE: proportion, 42.24 ± 6.27%), water-soluble species (36.60 ± 11.00%), organic matter (29.95 ± 3.42%, relatively wide range of 12.9%–45.7%), primary organic matter (single report, 34%), nitrate (17.01 ± 1.54%), and sulfate (16.15 ± 1.85%). Like the situation in Beijing, the studies indicate that vehicle emissions account for more than 30% of the $PM_{2.5}$ component of air pollution in Tianjin and Xi'an. The analogous portions in Chinese cities outside of Beijing (e.g., Shenzhen, Chengdu, and Huzhou) were lower (15%–27%). Our analysis indicates that in other Asian cities outside of China, the major source of $PM_{2.5}$ is coal combustion, and the principal pollutant constituents are EC, secondary OCs, and OAs. Their contributions vary dramatically across locations (Supplementary Table S6[1]).

According to our analysis, the sources of PM_{10} differed qualitatively across study sites in India, with crystal dust the main source in Delhi (49%–65%) and Tezpur (52%), OAs the main source in Shanghai (>50%), and road dust the main source in Kuala Lumpur (city center, 36%; suburbs, 55%) (see Supplementary Table S7[1] for detailed data). Meanwhile, we found that the pollutant species making up PM_1 in Beijing (Fig. 7*c*) were predominantly secondary products (72.50 ± 9.50%), followed by organic matter (40.0% ± 1.73%), nitrate (24.67 ± 2.03%), sulfate (17.67 ± 1.45%), and ammonium (16.00 ± 1.00%). Because there are relatively few articles about PM_1, it would be premature to draw strong conclusions. However, it is noteworthy that thus far there appears to be a substantial overlap between PM_1 constituents and $PM_{2.5}$ constituents, with the notable exception of less chloride in the former.

6. Summary and recommendations

In this article we present an overview of global research in air-SA techniques based on spatial and cluster bibliometric evaluation of research countries, research institutions, journals, key words, and research trends. During the study period (1980–2016), air-SA research sites shifted from the developed countries in Europe and North America to the developing countries in Asia. Methodologically, the common use of diffusion models waned in favor of receptor models; in recent years, combining online data monitoring (high temporal resolution) with various models has become popular. Over the study period, the main research objects shifted from coarse particles to fine and ultrafine particles. Aerosols have recently become important subjects of research, particularly in articles presenting methodological innovations. The meta-analysis revealed four directions in air-SA technology research: research for policy formulation, air pollution health-risk analysis, understanding the composition and formation of pollutants, and source apportionment improvement. Within Asia, China and India contributed the greatest volume of air-SA research, with Beijing and Delhi as common study sites and $PM_{2.5}$ and PM_{10} as common pollutant targets. Due to the lack of source inventory, among other reasons, PMF remains a mainstream method of analysis in Asia.

Using bibliometric and citation cluster analyses of publication outputs in air-SA techniques, this paper presents the spatial distributions and relationships of the countries and institutes featured and the temporal evolution of the knowledge clusters. The methods used in this work may be instructive in other research domains and locations, and they offer efficient and economic approaches to understanding research clusters and frontiers.

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