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## Journal Pre-proof

Source apportionment for indoor air pollution: Current challenges and future directions

Dikaia E. Saraga, Xavier Querol, Regina M.B.O. Duarte, Noel J. Aquilina, Nuno Canha, Elena Gómez Alvarez, Milena Jovasevic-Stojanovic, Gabriel Bekö, Steigvilė Byčenkienė, Renata Kovacevic, Kristina Plauškaitė, Nicola Carslaw



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## Source apportionment for indoor air pollution:

### Current challenges and future directions

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## **Abstract**

Source apportionment (SA) for indoor air pollution is challenging due to the multiplicity and high variability of indoor sources, the complex physical and chemical processes that act as primary sources, sinks and sources of precursors that lead to secondary formation, and the interconnection with the outdoor environment. While the major indoor sources have been recognized, there is still a need for understanding the contribution of indoor versus outdoor-generated pollutants penetrating indoors, and how SA is influenced by the complex processes that occur in indoor environments. This paper reviews our current understanding of SA, through reviewing information on the SA techniques used, the targeted pollutants that have been studied to date, and their source apportionment, along with limitations or knowledge gaps in this research field. The majority (78%) of SA studies to date focused on PM chemical composition/size distribution, with fewer studies covering organic compounds such as ketones,

carbonyls and aldehydes. Regarding the SA method used, the majority of studies have used Positive Matrix Factorization (31%), Principal Component Analysis (26%) and Chemical Mass Balance (7 %) receptor models. The indoor PM sources identified to date include building materials and furniture emissions, indoor combustion-related sources, cooking-related sources, resuspension, cleaning and consumer products emissions, secondary-generated pollutants indoors and other products and activity-related emissions. The outdoor environment contribution to the measured pollutant indoors varies considerably (<10%- 90%) among the studies. Future challenges for this research area include the need for optimization of indoor air quality monitoring and data selection as well as the incorporation of physical and chemical processes in indoor air into source apportionment methodology.

**Keywords:** indoor air quality, pollutants, source apportionment, receptor models

## Glossary

<b>AMS</b>	Aerosol Mass Spectrometry
<b>APCS</b>	Absolute Principal Component Scores
<b>CA</b>	Cluster Analysis
<b>CMB</b>	Chemical Mass Balance
<b>COA</b>	Cooking Organic Aerosol
<b>FA</b>	Factor Analysis
<b>IAQ</b>	Indoor Air Quality
<b>MLR</b>	Multilinear Regression
<b>OM</b>	Organic Matter

<b>(OP)FRs</b>	(Organophosphate) Flame Retardants
<b>PAEs</b>	Phthalate Esters
<b>PAHs</b>	Polycyclic Aromatic Hydrocarbons
<b>PCA</b>	Principal Component Analysis
<b>PM</b>	Particulate Matter
<b>PM<sub>2.5</sub></b>	Particles with a diameter of 2.5 micrometers or less
<b>PM<sub>10</sub></b>	Particles with a diameter of 10 micrometers or less
<b>PMF</b>	Positive Matrix Factorization
<b>PMF-ME</b>	Positive Matrix Factorization-Multilinear Engine
<b>PNC</b>	Particle Number Concentration
<b>PVC</b>	Polyvinyl Chloride
<b>QFASA</b>	Quantitative Fatty Acid Signature Analysis
<b>RM</b>	Receptor Model
<b>RPM</b>	Respirable Particulate Matter
<b>SA</b>	Source Apportionment
<b>SOA</b>	Secondary Organic Aerosols
<b>TSP</b>	Total Suspended Particles
<b>UFP</b>	Ultrafine Particles
<b>VOCs</b>	Volatile Organic Compounds
<b>WSOC</b>	Water Soluble Organic Compounds

## 1. Introduction

In the last two decades, a growing body of scientific evidence has shown that indoor air can be considerably more polluted than outdoors (Bekö et al., 2013). Different sources may contribute to the pollution in indoor environments, directly (e.g. emissions from consumer products, building materials, equipment, combustion sources, and infiltration of outdoor pollutants, (Weschler, 2009) or indirectly (i.e. through chemical reactions, (Weschler & Carslaw, 2018). Particulate and gaseous pollutants infiltrate indoors through convective flows (e.g. ventilation, open doors and windows) or diffusional flows/infiltration (e.g. cracks and fissures) (Roberts et al., 2023).

Human exposure to polluted air largely occurs indoors (Goldstein et al., 2021), given that people spend most of their time inside residences, offices, or other working microenvironments, public buildings or commuting. However, air quality standards and guidelines have been established primarily for outdoor air (Toyinbo et al., 2022). Considering the larger exposure time in different indoor microenvironments, the health effects of indoor air pollution - of both indoor and outdoor origin – are very relevant, especially among vulnerable population groups (Berglund et al., 1992; Kumar et al., 2023). Consequently, understanding the behavior and sources of indoor pollutants as well as distinguishing them from those originating outdoors is essential for both health risk assessment, establishing regulatory guidelines for indoor air quality (IAQ) and designing and applying mitigation strategies that aim to reduce human exposure to air pollution wherever it is received.

Source Apportionment (SA) is the practice of deriving information about the emission sources of air pollutants and their contributions to the overall air pollution. It includes a wide range of techniques used to obtain information about the influence that one or more sources have on a specific site or area over a specific time window. Such techniques may be based on the measured

concentrations of pollutants and their composition (receptor-oriented models) or on chemistry, transport and dispersion models (source-oriented models) (Belis et al., 2020). Focusing on receptor models, they can be classified into the following types: i) explorative methods that rely on empirical coefficients or diagnostic ratios between species, and ii) receptor models based on multivariate analysis of all the pollutant data at once (Hopke, 2016; Watson et al., 2008).

Receptor models apportion the measured mass of an atmospheric pollutant at a given site (receptor), to its emission sources by using multivariate analysis to solve a mass balance equation:

$$X = G F + E \quad (\text{Eq 1})$$

where  $X$  is a matrix containing ambient measurements of pollutant properties (typically chemical concentrations of gases and/or particles that include markers for different sources),  $F$  is a matrix whose rows represent the profiles (composition) of  $n$  sources,  $G$  is a matrix whose columns represent the contributions of the  $n$  sources and  $E$  is the residual matrix.

Those techniques applying weighted least-squares minimization fit (or other types of minimization techniques) to the ambient measurements using measured (a priori known) source profiles to solve the equation, are referred to as chemical mass balance methods (e.g., Chemical Mass Balance model, CMB). On the other hand, models that solve the equation without using 'a priori' information on source composition are factor analytical models, such as Positive Matrix Factorization (PMF) (Paatero & Tapper, 1994). Other receptor models include Principal Component Analysis (PCA), Factor Analysis (FA), UNMIX, Absolute Principal Component Analysis (APCA), Lanchow's approach, and Hybrid-trajectory-based models (Viana et al., 2008).

There is a continual improvement of SA tools in terms of source resolution and accuracy of source contribution quantification (Belis et al., 2015). A growing number of studies conducted for SA of outdoor air pollution have indicated that atmospheric concentrations at a specific location are affected by both local sources and regional background (D. Saraga et al., 2021), as well as a number of factors such as the landscape or the climatic and meteorological conditions (Karagulian & Belis, 2012; Viana et al., 2008). Focusing on particulate matter (PM) outdoors, a critical review and meta-analysis of SA studies in 20 European countries (Belis et al., 2013) resulted in six major source categories for PM: formation of secondary inorganic aerosol, traffic, re-suspension of crustal/mineral dust, biomass burning, (industrial) point sources, and sea/road salt. Studies on SA of outdoor Volatile Organic Compounds (VOCs), though more limited in number, indicate various anthropogenic sources, including industrial processes, solvent utilization, vehicle exhaust and gasoline evaporation, as well as biogenic sources (Sun et al., 2018).

Although the number of applications of SA for the outdoor air has been increasing steadily (Hopke, 2016), comparatively limited studies have attempted to identify and estimate the contribution of sources of indoor air pollutants using SA techniques. Source apportionment for indoor air is expected to differ from that for ambient air, mainly for the following reasons:

- There is significant variability within indoor environments, determined by the type and use of the buildings, the activities taking place in them, as well as the area where they are located.
- The multiplicity and high variability of indoor sources make it difficult to identify the sources with a potential for a significant contribution to the chosen SA model.

- The effects of air mixing and ventilation on pollutant transport, fate and formation in indoor atmospheres may influence allocation of sources inside a building.
- The role of the chemistry on indoor air pollutant dynamics. Homogeneous and heterogeneous (particularly important in the indoor environment due to the high surface to volume ratios) transformations can significantly influence pollutant concentrations (Bekö et al., 2020). Photochemistry and surface boundary layer reactions (e.g. indoor surfaces and deposited species are rapidly oxidized due to production of OH radicals within indoor boundary layer) are further factors that play a significant role (Morrison et al., 2019). Consequently, the assumption of stable source emissions (made by most of the SA models) needs to be applied with caution for indoor environments.
- Outdoor and indoor sources can be strongly interconnected (Gómez Alvarez et al., 2012). Some species originate from outdoors but can also be generated indoors, either by direct emission or via chemical transformation and secondary generation processes (e.g. ozone-initiated secondary organic aerosol formation enhanced by indoor precursors, (Nazaroff & Weschler, 2004).

All the above imply that SA of indoor air pollution is challenging. Indoor pollutant levels are controlled by both indoor and outdoor sources as well as by physical parameters (ventilation, light, temperature, relative humidity, outdoor conditions, etc.) that affect their physicochemical characteristics (Jia et al., 2019; Lakey et al., 2021; Sahu & Gurjar, 2021), at a spatial and temporal scale which is significantly different from outdoors. While the major indoor sources have been recognized, there is still need for understanding the contribution of the outdoor-generated pollutants penetrating indoors to indoor pollutant concentrations, as well as the role of the aforementioned parameters on SA.

The scope of this work is to present a review of the scientific literature on SA studies for indoor air between January 2009 and December 2022. A systematic search and review of peer-reviewed publications was used to gather information about the SA techniques used, the targeted pollutants, the identified sources and their contribution, as well as the effect of outdoor sources on indoor concentrations. In addition, this review identifies the limitations and knowledge gaps in this research field. As integrated information on the specific tracers for source identification become available, this review paper will serve as a guide for future SA studies for indoor air. To the best of our knowledge, this is the first time that an integrated literature review focuses on SA techniques applied to indoor air pollution.

## 2. Methods

A literature search covering SA for indoor air was conducted using the Google Scholar, Science Direct, Scopus and Web of Science search engines. Literature in the English language, published between January 2009 and December 2022 was included. The keywords used for the search were “indoor air” AND (“source apportionment” OR “receptor model” OR “positive matrix factorization” OR “chemical mass balance” OR “factor analysis” OR “principal component analysis” OR “enrichment factors” OR “diagnostic ratios” OR “time series analysis”).

To identify only literature relevant to the aims of this investigation, the following inclusion criteria were applied: the study (a) must contain indoor monitoring of air quality parameters and application of at least one SA method; (b) must have been conducted in a building; means of transport were excluded; and (c) studies including personal monitoring (at breathing zone) were only acceptable when indoor monitoring was simultaneously performed.

The search resulted in 251 papers. The titles and abstracts were screened by independent reviewers and 124 papers were removed for not meeting the above criteria. Finally, 127 papers were selected for further analysis. For each publication, the following information was obtained and categorized: (a) objective of the study; (b) detailed information on the type and number of indoor microenvironment(s), location of study; (c) targeted pollutant and tracers used (measurements and analytical techniques); (d) SA method; (e) output of the SA model (number and type of indoor and outdoor sources, quantified contribution of the sources); (f) potential physicochemical mechanisms influencing SA performance and results; and (g) potential limitations of the study.

### **3. Results and discussion**

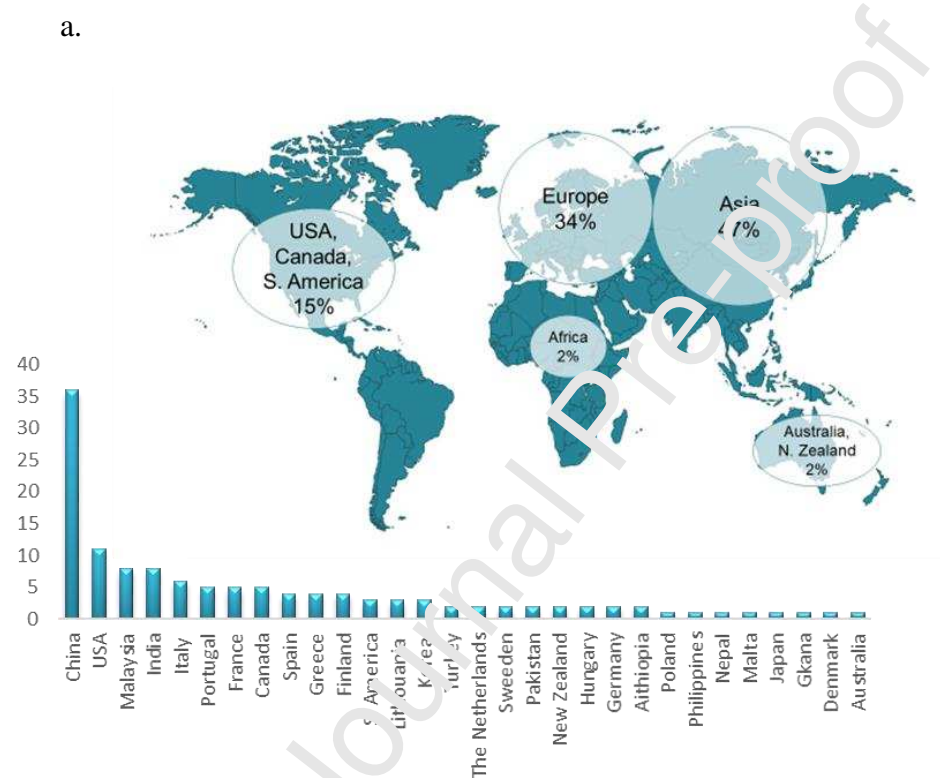
#### **3.1. Statistical overview**

According to the geographical distribution, 47% of the studies were conducted in Asiatic countries, 34% in Europe, 15% in USA, Canada and South America, 2% in Africa and 2% in Australia and New Zealand (Figure 1a). Regarding the indoor environment, 48% of the studies were conducted in residential buildings, 29% in schools and university buildings, and 11% in office buildings. In addition, 2% of the studies focused on elderly care home, 2% on restaurants and bars and 1% on hospitals. A limited number (7% in total) examined microenvironments with special characteristics (hotel, play center, industrial facility, church, and shopping mall). Thirty-six percent of the studies included sampling during two or more seasons. Forty-eight percent of the studies included coupled indoor and outdoor measurements, whilst 52% only covered indoor measurements.

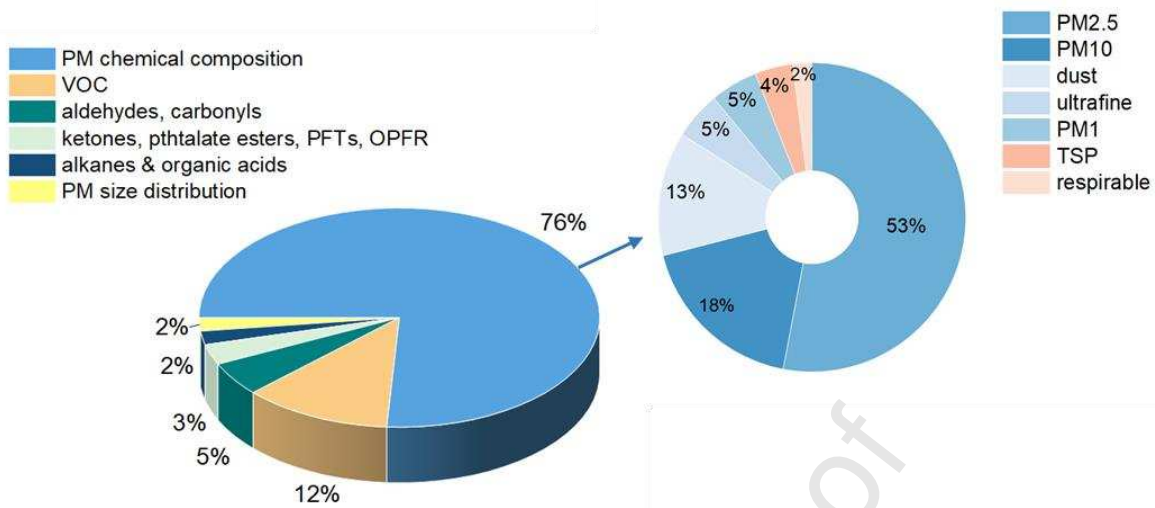
A fraction of the studies (10%) included monitoring at the occupants' breathing height (personal exposure), in addition to ambient monitoring at one or more indoor locations. Parallel personal, indoor and outdoor sampling is more time-consuming and labor-intensive compared to a fixed monitoring station study. Therefore, personal exposure datasets are often rather small (Molnár et al., 2014). Besides, the absolute (or relative) contribution of common indoor, ambient and personal sources may differ between the studies due to local and regional conditions, as well as the measured tracers used in the models, but it can be performed (e.g. Minguillón et al., 2012).

The overwhelming majority of the studies (78%) used PM (specific fractions or dust) chemical composition or PM size distribution as tracers to apportion measured concentrations to their potential sources. The remaining studies (22%) performed SA on VOCs, ketones, carbonyls and aldehydes, phthalate esters, PFTs or OPFR (Figure 1b). Regarding the SA method used, the majority of studies have used Positive Matrix Factorization (PMF) or Positive Matrix Factorization - Multilinear Engine (PMF-ME) (31%) Principal Component Analysis (PCA) (26%) and Chemical Mass Balance (CMB) (7 %) receptor models. Multilinear regression (MLR), Absolute Principal Component Scores (APCS), Factor Analysis (FA) and cluster analysis have been used in 1%, 3%, 3% and 2% of the studies, respectively. A non-negligible (21%) fraction of the publications have used more simple approaches (enrichment factors, diagnostic ratios, time series analysis, Spearman correlation) for SA purposes, occasionally combined with the application of a receptor model (Figure 1c).

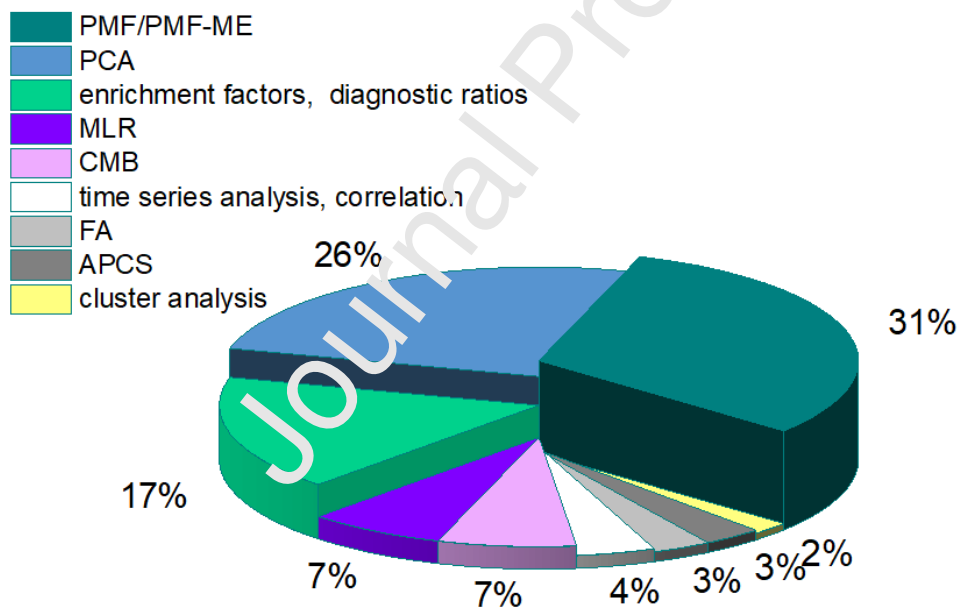
**Figure 1a-c.** Distribution of source apportionment studies (January 2009 – December 2022) for indoor air by a) number of studies per country b) targeted pollutants c) SA method used.



b.



c.



### 3.2 Methods used for indoor source apportionment

PMF, PCA, and CMB were used in 64% of the indoor SA studies. Studies that used the CMB model for indoor air SA were based on previous knowledge of the source profiles (Kopperud et al., 2004a; Lai et al., 2019), and the overwhelming majority of them focus on PM. The main advantage of the CMB model is that, unlike other statistical receptor models, it does not require a large input dataset (e.g., repeated measurements at a sampling location are not necessary). Moreover, unlike FA techniques, the CMB output does not require additional identification of the contributing sources/factors, as the profiles are selected a priori for well-defined sources. The model solves for the source contributions, while explicitly accounting for simultaneous indoor and outdoor sources of each species. On the downside, the lack of local (outdoor) or specific (indoor) source profiles data is a significant limitation of this approach. Even if available, the variation of the chemical profile of specific sources limits the application of receptor models, which need a priori knowledge of the source chemical profiles.

Studies using PMF and PCA methods for indoor SA are more abundant. They have been applied for source apportionment for both PM and VOCs. Their main strength is that the specific chemical profiles of the sources are not required. Thus, a priori selection of sources is not required, and the relevant ones are identified by multivariate analysis of the chemical datasets (PM or VOCs) obtained at the receptor. Another advantage is that by combining several microenvironments, all observations can be put into one large model. This can be important in studies involving time-consuming sampling strategies (e.g., off-line parallel outdoor, indoor, and personal exposure sampling and analysis), where the number of sampling days is a limiting

factor; or in large scale studies, where a large number of similar indoor environments (e.g. offices, schools) is targeted (Molnár et al., 2014; Campagnolo et al. 2017).

Furthermore, SA via multivariate models, e.g. PMF, might include both primary and secondary PM from each source, and for some source contributions it identifies processes rather than sources (i.e. secondary sulphate or nitrate factors) (Lai et al., 2019). Last but not least, in PMF, factor elements are constrained so that no sample can have a significantly negative factor contribution (non-negativity constraint). The lack of this constraint is a significant limitation of PCA and CMB.

However, there is a major limitation with regard to the application of PMF and PCA on indoor data. Indoor activities vary between buildings of different use (e.g. residential, office, school, hospital). Thus, the actual composition profile of indoor-generated pollutants is strongly variable from building to building, and even from day to day within one building. This variation does not comply with the basic assumption of PMF modeling that source profiles are stable over time, and such variation cannot be correctly represented by two or three factors (sources). Factor analysis-based approaches using personal exposure data are not free of this problem either. However, by using ME-2 or the last PMF version (v5 US-EPA), hybrid PMF-CMB analysis can be obtained by pooling and constraining specific emission chemical profiles that are known into PMF (Paatero & Tapper, 1994; EPA Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide).

In combination with receptor models, enrichment factor (EF) analysis has been used in several indoor SA studies, in order to estimate the contribution of the earth's crust and anthropogenic

activities to the PM elemental composition (Ali et al., 2017; Almeida-Silva et al., 2016; Gemenetzi et al., 2006; Lomboy et al., 2015; Mohamad et al., 2016; Zivkovic et al., 2022). An EF close to one indicates that the atmospheric element originated from natural or crustal sources while element EF values much higher than one suggest that the elements came from sea salt or anthropogenic sources, such as combustion, vehicle and industrial emissions. Al and Ti are typically used as reference elements for EF, since construction works can affect the content of Si, K, Ca, road dust can affect Fe, Mn, Ca, biomass burning can affect K and sea salt can affect Na. IAQ measurements have been used by other than receptor models, which are mainly used to predict indoor air concentrations over a wide range of conditions and to provide insight into the underlying physicochemical processing (Shiraiwa et al., 2019). These models are not typically used for SA and are not discussed in this paper.

### **3.3 Target pollutants**

#### **3.3.1. Particulate matter**

The most frequently used PM fraction in indoor SA studies is fine particulate matter,  $PM_{2.5}$ , followed by  $PM_{10}$ ,  $PM_{10-2.5}$  and ultrafine particles (UFP). Fewer studies have focused on total suspended particles (TSP), settled dust on indoor surfaces and respirable PM or  $PM_4$  (Figure 1b). Since outdoor  $PM_{2.5}$  and  $PM_{10}$  are regulated [e.g. European Directive 2008/50/EC (EN 12341 and EN 14907) ; U.S. EPA NAAQS], these fractions are more extensively monitored; consequently, indoor air SA studies in the last decade have mainly focused on these fractions.

The SA methods have been applied either on the chemical composition of the particles or on their size distribution. The former approach includes impactor sampling (gravimetric filter-based

method) and off-line analysis in the laboratory for major chemical components as ions, metals, organic/elemental/black carbon, polycyclic aromatic hydrocarbons (PAHs), water-soluble organic compounds (WSOC), levoglucosan and organic acids (Bergmans et al., 2022; Duarte et al., 2022). The second approach uses particle number concentration (PNC) and particle number size distribution (PNSD). Ultrafine particles are often measured in indoor studies because of their high health impact potential, and the fact that indoor emissions and aerosol processes might have a larger impact on PNC than on particle mass concentrations. However, the number of indoor studies on SA of size-resolved UFP is relatively scarce. (Bellö et al., 2013) used diary entries regarding occupancy and particle related activities to apportion the daily integrated PM exposure to individual sources in 56 Danish residences. In a more recent study, Ouaret et al., 2021 applied the non-negative Matrix Factorization method on continuous (resolution of 1 min) PM size distribution (0.3 - 20  $\mu\text{m}$ ) data collected in an occupied workspace over a six-month period. The apportionment results were expressed as source diurnal profiles (temporal fingerprints) and the obtained source contributions were related to the office occupancy and natural ventilation patterns. In another recent SA study, a novel multi-heights sampling scheme using low-cost sensors was used for estimating the contribution of cooking to  $\text{PM}_{2.5}$  levels inside an urban apartment in Beijing (Aliwaer et al., 2022). Continuous (minute scale) PM measurements at eight heights in the kitchen and bedroom revealed a strong vertical variation in the kitchen, that was only apparent during the cooking events. In contrary to the one-location sampling, the proposed scheme can be applied to characterize the vertical profiles of air pollutants in a wide range of indoor environments, with different ventilation systems and emission sources.

### 3.3.2. Organic compounds

Over 20% of the studies included in this review performed indoor SA on VOC, carbonyls and aldehydes (Figure 1b). The identification of indoor sources of these organic compounds is challenging, as many of these species are emitted by multiple sources in both indoor and outdoor environments (Spinazzè et al., 2020). Furthermore, the analytical approaches to measure these organic compounds indoors are resource demanding. However, due to the health effects of indoor exposure to VOCs, investigating the origin and behavior of VOCs in various micro-environments is of utmost importance.

Indoor VOCs, carbonyls and aldehydes can originate from a wide variety of sources, the strength of which may vary depending on the season (i.e. differences in temperature, relative humidity and other environmental conditions), as highlighted by several SA studies (e.g. Huang et al., 2018; Jia et al., 2008). Normally, SA studies of indoor VOCs, carbonyls and aldehydes use time averaged samples and use multivariate receptor models. These analyses typically use data reduction techniques (PCA or PMF) on population level data acquired in survey studies (Campagnolo et al., 2017). Nevertheless, given the nature of indoor VOC, future studies should focus on a comprehensive profile of these organic compounds at different indoor sites to allow a better understanding of the identity and strength of the indoor sources in different locations.

### 3.4 Identified sources and contributions

The number of sources identified in each indoor SA study ranges between 2 and 13. The overwhelming majority of the studies highlight the contribution of outdoor sources (1 to 7 specific outdoor sources have been identified). Nevertheless, the number and type of identified sources is strongly dependent on the selection of the chemical species used as tracers for the SA

methods. For example, it is impossible to identify the secondary organic PM contribution if specific organic tracers are not analyzed, or shipping emissions without the analysis of the chemical elements vanadium (V) and nickel (Ni).

The indoor sources considered in the SA literature can be grouped as follows: i) building materials and furniture ii) combustion-related, iii) cooking-related, iv) resuspension, v) cleaning and consumer products, vi) indoor generated secondary pollutants and vii) other products and activities. In approximately one third of the studies, two or more sources were presented as combined or mixed sources, as the model did not permit them to be distinguished from each other. This usually happens when the component used as a tracer corresponds to more than one group of sources. Table 1 summarizes the resolved sources, their tracers and the SA method used, as derived from the present literature review. Furthermore, Figure 2a,b presents the percentage contribution of specific indoor sources to PM and VOCs, as derived from the SA studies review. A comparison between percentages should be done with caution, as different approaches -in sampling, analysis and modeling- may have been used.

**Table 1.** Identified sources, their tracers and the SA method used

Source Group	Source characterized by the SA studies	Tracers	SA method	Reference
Crustal sources & resuspension	Resuspension/dust/crustal	Ca, S, Al, Fe, Mg, Mn, Cr, Cu, Zn, Ba, Ni, Ti, Rb, Li	PMF, PMF-ME	Alias et al., 2021; Amato et al., 2014; Carrion-Matta et al., 2019; Molnár et al., 2014; Niu et al., 2021; Suryawanshi et al., 2016; Tunno et al., 2016
			Mass Balance	Kopperud et al., 2004

			PCA	Mohamad et al., 2016; Taner et al., 2013
			EF	Taner et al., 2013
<b>Indoor combustion sources</b>	<b>Smoking/Environmental Tobacco smoke</b>	Ni, V, Mg, Ca,Cd, Cu, K,Tl, As, La, Ce/NO <sub>2</sub> / Organic carbon, Elemental carbon, Black Carbon/ benzo(k)fluoranthene, benzo(ghi)perylene/ toluene, ethylbenzene, benzene, 1,4-cichlorobenzene, styrene, hexane, cyclohexane, heptane, nonane, 1,3,5-trimethylbenzene, decane/nicotene	PMF, PMF-ME, Varimax Rotated Factor Analysis	Bari et al., 2015; Barraza et al., 2014; Basaran & Yilmaz Civan, 2021; Clougherty et al., 2011; Huang et al., 2019; Minguillón et al., 2012; Pekey et al., 2013; Saraga et al., 2010; Shang et al., 2019; Suryawanshi et al., 2016; Tunno et al., 2016; Yang et al., 2018
			PCA	Romagnoli et al., 2016
			EF, LUR, Multivariate Curve Resolution Alternating Least Squares, diagnostic ratios	Clougherty et al., 2011; Romagnoli et al., 2016; van Drooge et al., 2018; Yang et al., 2018
	<b>Biomass/wood burning/ charcoal combustion (indoors)</b>	K <sup>+</sup> , K, S, Cl, Zn, Br, As, Se, V/Organic carbon, Elemental carbon, Black Carbon/ benzo(a)anthracene, acenaphthylene, benzo(b)fluoranthene, benzo(a)pyrene phenanthrene, benzo(e)pyrene, benzo(k)fluoranthene benzo(ghi)perylene, anthracene, fluoranthene, pyrene, chrysene/ VOC, WSOC, levoglucosan, mannosan, galactosan, glucose, mannitol/soot	PMF, PMF-ME	Adeniran et al., 2021; Basaran & Yilmaz Civan, 2021; Canha et al., 2014; Minguillón et al., 2012; Zhou et al., 2014; Zhu et al., 2012
			CMB	Lai et al., 2019
			PCA	Liu et al., 2022; Taner et al., 2013
			EF, diagnostic ratios,MLR	Adeniran et al., 2021; Canha et al., 2014; Y. Liu et al., 2022; Taner et al., 2013

	<b>Coal combustion (indoors)</b>	Pb, As, V, Se, Cs, Cl <sup>-</sup> / benzo[ghi]perylene	PMF	Rybak et al., 2019; Shang et al., 2019
			PCA	Taner et al., 2013
			EF, cluster analysis, diagnostic ratios	Rybak et al., 2019; Taner et al., 2013
	<b>Incense burning</b>	ultrafine particles; acenaphthylene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(e)pyrene, benzo(a)pyrene, benzo[ghi]perylene	PCA	Masih et al., 2012
			time series analysis	Bekö et al., 2013
	<b>Combustion processes</b>	butanal, isobutanal, acetone, acetaldehyde, methyl vinyl ketone (MVK), 2-Methyl-2-propenal, acrolein, acetonitrile, 2-methylfuran, isoprene, methylacetate	PMF	Bari et al., 2015
<b>Cooking</b>	<b>Cooking (general)</b>	As, Zn, Ca, Mn, Fe, Cd, Cu / formaldehyde, o-xylene, styrene, benzene, 1,2,4-trimethylbenzene, benzyl chloride, trichloroethane, tetrachloroethane / ethanol, methanethiol, pyrrole, dimethyl sulfide, diethyl (1,1)-but-2-enedithiolate, hydron (C6H9O4+), D4 siloxane, D5 siloxane, dimethoxysilane/ benzo(a)pyrene, acenaphthene, phenanthrene, benzo[ghi]perylene, benzo(b)fluoranthene, fluoranthene, indentalene, benzo(a)anthracene, chrysene, benzo(k)fluoranthene, benzo(e)pyrene /WSOC	PMF	Adeniran et al., 2021; Basaran & Yilmaz Civan, 2021; Huang et al., 2019; Katz et al., 2019; Niu et al., 2021; Rybak et al., 2019; Tunno et al., 2016
			CMB	Arhami et al., 2010
			PCA, APCS	Masih et al., 2012; Ohura et al., 2009; Othman et al., 2016; Taner et al., 2013
			EF, MLR, diagnostic ratios, time series analysis, Varimax Rotated Factor Analysis,	Adeniran et al., 2021; Lunderberg et al., 2021; Othman et al., 2016; Taner et al., 2013; Yang et al., 2018
	<b>Chinese cooking</b>	oleic acid, mannosan, galactosan	CMB	Lai et al., 2019
	<b>Meat cooking</b>	As, V, Se, Cr, Cu, Ni, Zn	PCA, EF	Taner et al., 2013
<b>Kitchenware/ stainless steel utensils</b>	Ni, Cr, Fe, Cu	PMF, PCA, EF	Niu et al., 2021; Taner et al., 2013	
<b>Cooking oil fumes</b>	Zn, Cu, Ca/Fatty acids: C18:1 n-9, C18:2 n-6, C20:1 n-9/ 2-methylnaphthalene, 1-methylnaphthalene, biphenyl, carbazole	PMF, PCA, MLR, EF	Masih et al., 2012; Othman et al., 2016; Taner et al., 2013; Xie et	

				al., 2021
<b>Building materials &amp; furniture</b>	<b>Building materials (general)</b>	Al, Pb, Cu, Zn/benzyl butyl phthalate, di-n-octyl phthalate/ VOCs (isopropyl benzene, p-isopropyl toluene, styrene, phenol, d-limonene), heavy alkanes	PMF	Li et al., 2021; Niu et al., 2021
			PCA, APCS, FA, MLR, EF	Ali et al., 2017; Jia et al., 2010; Jung et al., 2021; Li et al., 2021
	<b>Off-gassing of building materials</b>	hexanal, butyl acetate, formaldehyde, 2-heptanone, 1-butanol, cyclohexanone, benzaldehyde hexanal, butyl acetate, benzaldehyde/ Acetone, acetaldehyde, glyoxal, benzaldehyde, glyoxal, methyl isobutyl ketone/ pinene	PMF	Bari et al., 2015; Y. Huang et al., 2019
			PCA, APCS	Guo, 2011
			time series analysis	Lunderberg et al., 2021
	<b>Aging of materials</b>	Pentanal, hexanal, octanal, nonane	PCA, NMF	Rosch et al., 2014; Ali et al., 2017; Jung et al., 2021
	<b>Corrosion of building facades/windows frames</b>	Al, Cd	PCA, APCS, FA, MLR, EF	Ali et al., 2017; Jung et al., 2021
	<b>Flooring emissions (general)</b>	benzene, ethyl benzene, styrene, toluene, 1,2,4-Trimethylbenzene, 1,3,5-Trimethylbenzene, m,p-xylene,o-xylene,n-hexane, n-nonane, n-undecane, n-tetradecane, n-hexadecane, limonene, $\alpha$ -pinene, $\beta$ -pinene, 2-butanone, p-dichlorobenzene/ decane, nonane, undecane, dodecane, trimethylbenzene isomers, n-butylbenzene, n-propylbenzene, ethyltoluene isomers, octane/ 2-butoxyethanol, 2-ethylhexanol, propanal	PMF, PCA	Bari et al., 2015; Campagnolo et al., 2017; Shin & Jo, 2012
	<b>PVC flooring</b>	Tetrahydrofuran	PCA, NMF	Rosch et al., 2014
	<b>Wood/ particle board flooring</b>	C12-C16 alkanes/1,4-di-chlorobenzene/ n-DDC, n-TTD, n-PTD, $\beta$ -PN, NNA/ formaldehyde/ terpenes, 2-heptanone	PMF, PCA	Y. Huang et al., 2019; Q. Liu et al., 2014; Shin & Jo, 2012
<b>Wall covering</b>	Ca, Cu, Fe, Pb, Mg, Ni/ Toluene, trimethylbenzenes/n-heptane, n-octane, n-nonane	PMF	L. Huang et al., 2018; Suryawanshi et al., 2016	
<b>Ceiling - mineral fiber tiles/ melanine foam</b>	CH <sub>2</sub> O	Emission rates measurements	Poulhet et al., 2014	
<b>Wood furniture</b>	n-butyraldehyde/ terpenes/ formaldehyde/ styrene/n-butanol/ Decan, un-, do-, tridecan, pinene/hexanal	PMF	L. Huang et al., 2018; Suryawanshi et al., 2016	
		CMB	Plaisance et al., 2017	
		PCA, NMF	Rosch et al., 2014; Campagnolo et	

				al., 2017
	<b>Waxes</b>	Palmitoleic, oleic acids	Molecular marker chemical mass balance model)	Daher et al., 2011
	<b>Steel pipes</b>	Mn	PCA, APCS, MLR, EF	Ali et al., 2017
	<b>Insulation materials</b>	PBDEs	PMF	Basaran & Yilmaz Civan, 2021
	<b>coatings (polystyrene, thermoset resin, polycarbonate, thermoplastic)</b>	1,2-Bis(2,4,6-tribromophenoxy)ethane, tetrabromophthalate, Zn	PMF, PCA, EF	Canha, , et al., 2014b; Yadav & Devi, 2022
	<b>latex</b>	acrolein, benzaldehyde	PCA	Campagnolo et al., 2017
<b>Consumer products &amp; materials</b>	<b>Household products (general)</b>	ethanol, methylene chloride, acetone, acetaldehyde, chloroethane, 1,1-dichloroethane, cyclohexane/ ethylbenzene, o-, m- and p-xylene toluene, o-xylene, TVOC, WSOC	PMF	Bari et al., 2015; Y. Huang et al., 2019
			PCA, APCS, diagnostic ratios	Ohura et al., 2009; Romagnoli et al., 2016
			molecular marker-based chemical mass balance (MM-CMB)	Hasheminassab et al., 2014
	<b>Insect repelen, pesticide, Camphor</b>	1,4-Dichlorobenzene, hexachlorobutadiene, 1,1,2,2-tetrachloroethane, trans-1,3-dichloropropene, 1,1-dichloroethane, trichloroethylene, cis-1,2-Dichloroethylene/ Nap	PCA, FA	Bai et al., 2022; Jia et al., 2010
	<b>Solvent use</b>	ethanol, acetone, hexane, heptane, methylcyclopentane, cyclohexane, BDE-28, BDE-47, BDE-99, PBEB, hexachlorobutadiene, 1,1,2,2-tetrachloroethane, tetrachloroethylene, chloropropene, carbon tetrachloride, benzene, 1,1,2-trichloro-1,2,2-Trifluoroethane	PCA, APCA, NMF, spearman correlation	Rosch et al., 2014; Bai et al., 2022; Widiana et al., 2017; Yadav & Devi, 2022
	<b>Polish remover</b>	acetone	time series analysis	Lunderberg et al., 2021
	<b>Cosmetics , personal care products</b>	dimethyl phthalate (DMP), diethyl phthalate (DEP)/ methyl-dihydrojasmonate , galaxolide / Ca, Li, Ti, Sr, Cyclic siloxanes	PMF, PCA. time series analysis	Li et al., 2021; Lunderberg et al., 2021; Martins et al., 2016; Minguillón et al., 2012

<b>Fragranced consumer products, deodorizers, fresheners</b>	limonene, a-pinene, b-pinene, camphene, p-cymene, styrene, ethylacetate, dodecane, p-DCB, naphthalene, TCA, 1,2,4-trichlorobenzene, dichlorobenzene, cis-1,3-dichloropropene, 1,1,1-trichloroethane, trichloromethane, benzyl chloride, 1,2-dibromoethane, tetrachloroethylene, trans-1,3-dichloropropene, 1,2-dichloropropane, trichloroethylene, 1,1-dichloroethylene	PMF, PCA, APCS	Bai et al., 2022; Bari et al., 2015; Guo, 2011
<b>Cleaning products, detergents, additives</b>	benzene, toluene, ethylbenzene, xylenes, chlorobenzene, 1,2,4-trichlorobenzene, halogenated hydrocarbons, 1,2-dibromoethane, cis-1,3-dichloropropylene and trichloroethylene/ dichloroamine/ P, Cl- /2-butoxyethanol	PMF	Barmeparesos et al., 2020; Barraza et al., 2014; Q. Liu et al., 2014
		PCA, FA	Bai et al., 2022; Clougherty et al., 2011; Campagnolo et al., 2017
		time series analysis	Lunderberg et al., 2021
<b>Paints (water-based latex paints, solvent-based paints, decorative arts)</b>	Pb, Cu, Cr / toluene, trimethylbenzenes/n-heptane, n-octane, n-nonane, 4-methyl-2-pentanone, n-butyl acetate, tetrachloroethylene/ 1,3,5-TMB, FORM, BACT, 1,2,4-trichlorobenzene, 1,2,4-trimethylbenzene, 1,2-dibromoethane, chloropropene and 1,2-dichloropropane	PMF	L. Huang et al., 2018; Q. Liu et al., 2014
		PCA, APCS, MLR, EF, FA	Ali et al., 2017; Bai et al., 2022; Jia et al., 2010; Jung et al., 2021; Shin & Jo, 2012
<b>Adhesives</b>	2-Propanol, 2-Butanone	PCA	Shin & Jo, 2012
<b>Paints &amp; adhesives (general)</b>	benzene, toluene, ethylbenzene, xylenes/heptane, methylhexane, methylcyclohexane, dimethylhexane methyl, ethyl ketone, methyl isobutyl ketone/ methyl butyl ketone, acetone, 1,2-dichloroethane, methyl isobutyl ketone, benzyl chloride/ MVK, acetone, 1,2-dichloromethane, MIBK, benzyl chloride/ Tri-, tetra-, penta-, hexadecane	PMF, PCA, APCS	Bari et al., 2015; L. Huang et al., 2018; Y. Huang et al., 2019; Ohura et al., 2009
<b>Chalk</b>	Ca, Ca <sup>+</sup> , Sr	PMF, PCA, APCS, EF	Amato et al., 2014; Canha et al., 2014a,b; Mohamad et al., 2016
<b>PVC, polystyrene, polyamides</b>	BDE-154, BDE-209, syn-DP, anti-DP, TMPP	PMF, PCA	Yadav & Devi, 2022
<b>Plasticizers employed in multifarious plastic</b>	DIBP, DBP, DEHP, DNP	PMF, PCA	Li et al., 2021

	<b>Paper products (dye and paper manufacturing)</b>	OPAH 9,10-anthraquinone, 1,1,2-trichloroethane, toluene, 1,1-dichloroethane, benzene, 1,2-dichloroethane, 1,1,1-trichloroethane and cis-1,2-dichloroethylene	PMF, PCA, diagnostic ratios	Bai et al., 2022; Lim et al., 2021
<b>Other sources</b>	<b>Home appliances, computer cases</b>	deca-BDEs, Octa-PBDEs, penta-PBDE	PMF	Basaran & Yilmaz Civan, 2021
	<b>Steel-making</b>	Fe, Mn, Pb, Zn	PMF	Tunno et al., 2016
	<b>Domestic wastewater decomposition</b>	H <sub>2</sub> S, NH <sub>3</sub>	PCA, APCS	Widiana et al., 2017
	<b>Chlorination treatment process</b>	Methylene chloride	PCA, APCS	Widiana et al., 2017
	<b>Tap water and bleach use</b>	halogenated VOCs (Freon compounds, carbon tetrachloride, trichloroethylene, 1,1,1-trichloroethane, bromomethane, chloromethane, p-dichlorobenzene), carbon disulfide, chloroform, dichloroacetate, dichloromethane, perchloroethylene	PMF	Bari et al., 2015
	<b>Clothes</b>	Fatty acids: C18:1 n-12, C16:1T, C12:0	PCA	Xie et al., 2021
	<b>Pet hair</b>	Fatty acids: C18:0, C18:1 n-12, C24:0	PCA	Xie et al., 2021
	<b>Human hair</b>	Fatty acids: C14:0, C15:0, C16:0, C13:1, C17:0, C18:2T, C13:0	PCA, Molecular marker chemical mass balance model	Daher et al., 2011; Xie et al., 2021
	<b>Dandruff</b>	Fatty acids: C15:0, C16:0, C17:0, C18:2T	PCA	Xie et al., 2021
		<b>Fireworks</b>	K, Sr, Ba	PMF, MLR
	<b>lighters</b>	Ce	PMF	Minguillón et al., 2012
<b>Outdoor sources (resolved from indoor SA studies)</b>	<b>Traffic</b>	organic carbon, elemental carbon, black carbon/PAHs (fluoranthene, pyrene, benzo(a)anthracene, chrysene)/benzene, toluene, o-, m-, p- xylene/ ethyltoluene isomers, trimethylbenzene isomers, propylbenzene, ethylbenzene, Zn, Cr, Ni, As,	PMF	Bari et al., 2015; Basaran & Yilmaz Civan, 2021; Carrion-Matta et al., 2019; Pekey et al., 2013
			PCA, hierarchical cluster analysis (HCA), polytopic vector analysis (PVA), cluster analysis	Al-Harbi et al., 2020; Bravo-Linares et al., 2016
	<b>Exhausts (diesel, gasoline)</b>	NO <sub>3</sub> <sup>-</sup> , NO <sub>2</sub> , SO <sub>2</sub> /organic carbon, elemental	PMF	Adeniran et al.,

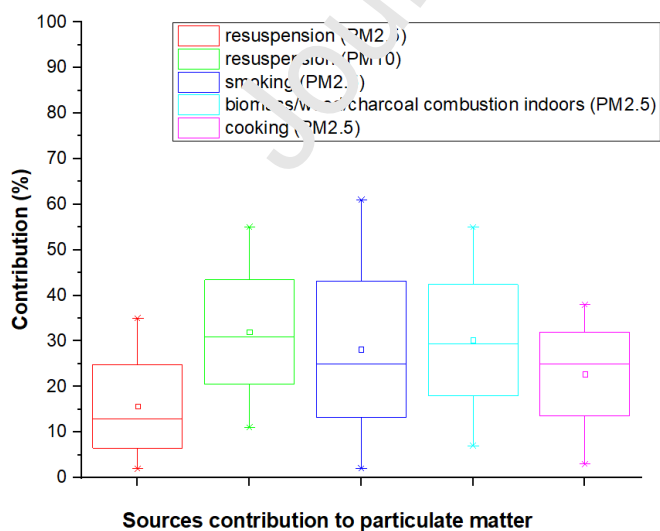
		carbon/ benzene, cyclohexane, heptane, toluene, octane, ethylbenzene, xylenes, styrene, nonane, 1,3,5-trimethylbenzene, decane/ benzo(a)pyrene, chrysene, benzo(b)fluoranthene, benzo(ghi)perylene, fluoranthene, pyrene, benzo(a)anthracene, benzo(k)fluoranthene		2021; Heo et al., 2021; Pekey et al., 2013
			PCA, diagnostic ratios	Adeniran et al., 2021; Cao et al., 2019
<b>Biomass/wood burning</b>		PAHs/ organic carbon, elemental carbon, black carbon, brown carbon /K, Zn, Pb/levoglucosan, water-soluble K, WSOC	PMF	Carrion-Matta et al., 2019; Fernandes et al., 2021; Ji et al., 2022
			CMB	Lai et al., 2021
			PCA, polytopic vector analysis (PVA), EF, spearman correlation, time series analysis	Bravo-Linares et al., 2016; Fernandes et al., 2021; Jeong et al., 2019; Ji et al., 2022; A. Lai et al., 2021; Pauraite et al., 2021
<b>Soil</b>		Al, Ca, Mg, Mn, K, Ce, Eu, Fe, Ti, Hf, La, Li, Rb, Sr, S, Th, Sm, Co, Sc/ Organic carbon/ Al <sub>2</sub> O <sub>3</sub> / Ca <sup>2+</sup> , Mg <sup>2+</sup>	PMF	Heo et al., 2021; Ji et al., 2022; Minguillón et al., 2012; Niu et al., 2021; Shang et al., 2019
			CMB	Lai et al., 2021
			PCA, HCA, EF, diagnostic ratios, spearman correlation, Varimax Rotated Factor Analysis	Al-Harbi et al., 2020; Almeida-Silva et al., 2016; Canha, Almeida, Freitas, Trancoso, et al., 2014; Ji et al., 2022; A. Lai et al., 2021; Yang et al., 2018
<b>Road dust/brake/tyre wear</b>		Pb, Al, Ca, Se, Mg, Si, Fe, S, BC, Cr, Mo, Sb, Cu, Zn, Mn, BS/ phenanthrene, anthracene	PMF	Brehmer et al., 2019; Tunno et al., 2016
			PCA, APCS, MLR, EF, HCA	Ali et al., 2017; Fernandes et al., 2021; Habil et al., 2013;

				Jeong et al., 2019; Mohamad et al., 2016; Taner et al., 2013
	<b>Sea salt</b>	$\text{Na}^+, \text{Cl}^-, \text{SO}_4^{2-}, \text{Mg}^{2+}, \text{Ca}, \text{Mg}, \text{K}, \text{Na}, \text{Cl}$	PMF	Carrion-Matta et al., 2019; Heo et al., 2021; Ji et al., 2022
			PCA, EF	Almeida-Silva et al., 2016; Canha, Almeida, Freitas, Trancoso, et al., 2014; Ji et al., 2022
	<b>Secondary aerosol, secondary pollution</b>	$\text{SO}_4^{2-}, \text{NO}_3^-, \text{NH}_4^+$	PMF	Carrion-Matta et al., 2019; Heo et al., 2021
			CMB	Arhami et al., 2010
			PCA	Almeida-Silva et al., 2016; Canha et al., 2014a
	<b>Long-range transport</b>	S, S <sub>o</sub> , Pb, Mn	FA	Clougherty et al., 2011
	<b>Industrial emissions</b>	organic carbon, elemental carbon, $\text{SO}_4^{2-}$ , $\text{NO}_2$ , SO, Fe, Zn, Mn, Pb, Ba, Co, As, Cr, Cu, Ni, S, V / benzene, toluene, ethylbenzene, xylenes / methyl-pentane, dimethylbutane, hexane, isopentane, methyl-cyclopentane, cyclopentane, pentane	PMF	Bari et al., 2015; Heo et al., 2021; Minguillón et al., 2012; Pekey et al., 2013
			PCA, HCA,	Al-Harbi et al., 2020
	<b>Fuel oil combustion</b>	organic carbon, elemental carbon, black carbon/ $\text{SO}_4^{2-}$ , $\text{NO}_3^-$ , /Fe, V, Ni, V, Ca, Na, Mg, Ni, Cu, Pb /trans-2-pentene, cis-2-butene, trans-2-butene, 1-pentene, 2-methyl-2-butene, 3-methyl-1-butene, 2,2,4-trimethylpentane, 2,4-dimethylpentane, 2,3,4-trimethylpentane, toluene, butane, isopentane, pentane, benzene, 2,3-Dimethylbutane	PMF	Bari et al., 2015; Fernandes et al., 2021; Heo et al., 2021; Minguillón et al., 2012; Niu et al., 2021
			PCA, APCS, MLR, EF, FA, spearman correlation	Ali et al., 2017; Clougherty et al., 2011; Fernandes et al., 2021
	<b>Oil and gas industry</b>	ethane, propane, butane, isobutane, ethylene, acetylene, 1,3-butadiene, propene, propyne, 1-butene, benzene, cyclohexane, 2,2-dimethylbutane, hexane, pentane and isopentane/ 2- to 3-ringed PAH	PCA, polytopic vector analysis (PVA), spider webs	Bari et al., 2015; Bravo-Linares et al., 2016; Rybak et al., 2019

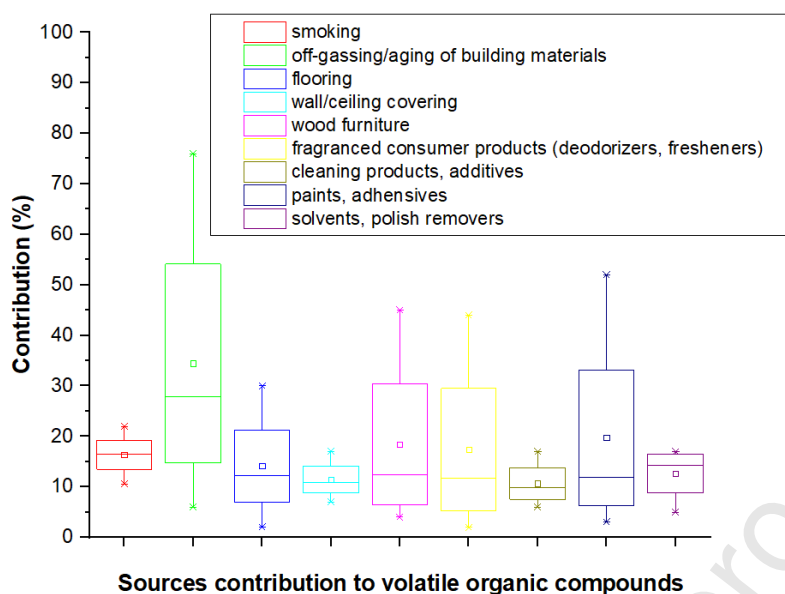
	<b>Unburnt fossil fuels (lubricating oil combustion, kerosene, natural gas combustion-LNG, aerosol sprays (petrochemicals), and unburnt diesel and gasoline)</b>	naphthalene, acenaphthene, benzo(a)anthracene	PMF, diagnostic ratios	Adeniran et al., 2021
	<b>Solid waste/garbage combustion</b>	organic carbon, elemental carbon, $K^+/K$ , Cu, Cd, Ni	PMF, CMB, PCA	Bano et al., 2018; Habil et al., 2013
	<b>Coal combustion</b>	Fe, Pb, Ba, Br, Ca, Mg, As, Cs, S, Se, Cl/phenanthrene, anthracene	PMF	Suryawanshi et al., 2016; Tunno et al., 2016
			PCA, diagnostic ratios, FA	Cao et al., 2019; Clougherty et al., 2011
			CMB	Lai et al., 2021

**Figure 2a, b.** Box plots for indoor sources contribution to a) PM and b) VOCs. Source types identified in more than five studies were included in the statistical analysis. Combined sources identified as one source were excluded from the analysis.

a)



b)



### 3.4.1 Building materials and furniture emissions

This group of sources includes emissions from paints, solvents, adhesives, particle board and plywood materials, other wood-based materials and wall coverings. SA studies indicate that their contribution to the targeted pollutants concentrations (which are mainly organic compounds) ranges between 10% and 75% (Figure 2b).

The process in which household materials (e.g. particle-board, fiberboard, floor lacquers, molded plastics, adhesives) release VOCs into the air, known as off-gassing, has often been identified as a distinct indoor source, with a remarkably different contribution among the SA studies. For instance, the off-gassing from building materials has been identified as the prevailing source of TVOC (76%, traced by formaldehyde and pinene) by (Guo, 2011) who applied PCA and APCS models on data from 100 Hong Kong residences. The remaining 24% was attributed to household products, painted wood, room freshener and mothball. In contrast, (Bari et al., 2015) who applied PMFv.3 on polar and non-polar VOCs from 50 Canadian houses, attributed only 6%

of the TVOC to off-gassing from building materials (traced by hexanal, butyl acetate, formaldehyde, 2-heptanone, 1-butanol, cyclohexanone, benzaldehyde) while emphasizing the importance of emissions from paints and adhesives (traced by heptane, methylhexane, methylcyclohexane, dimethylhexane methylethylketone, methyl isobutyl ketone) and floor/wall coverings (traced by decane, nonane, undecane, dodecane, trimethylbenzene, n-butylbenzene, ethyltoluene, octane). These substantial differences between the studies likely reflect the selection of tracers rather than the differences in building age and composition.

Lunderberg et al., 2021 performed SA on multi-season time series of 200 VOCs measured by a fast-response online mass spectrometer in two Californian residences. Although VOC emissions from buildings are expected to be less important in older buildings, they observed that continuous indoor sources were the largest contributors to VOC for more than 90% of observed species, even though the residences were decades old and had not been recently remodeled or refurnished. These indoor sources included i) continued material off-gassing as deeply bound VOCs migrate to surfaces; ii) oxidative, thermal, or microbial decomposition of building materials yielding decomposition products; or iii) sorptive interactions as semi-VOC or VOC related to occupant activities deposit on surfaces and then slowly off-gas. This latter phenomenon may cause primary emissions from episodic sources to be interpreted as originating from building materials, influencing the SA results.

SA studies attribute a substantial part of indoor VOC to building materials and furniture emissions. (L. Huang et al., 2018) used PMT to attribute 4 out of 7 VOCs sources in 27 residences in Beijing to building materials and furniture emissions. Emissions from hydrolysis of resins in building materials and furniture were traced by n-butyraldehyde, whilst wood-flooring emissions were characterized by high loadings of 1,4-dichlorobenzene and long-chain alkanes,

which are common ingredients of wood varnishes. Emissions from wooden furniture were traced using terpenes and formaldehyde, whilst emissions from wall coverings were linked to toluene, trimethylbenzenes, n-octane and n-nonane, which are common components of water-based latex paints and solvent-based paints.

In a relative PMF study, (Y. Huang et al., 2019), attributed 45% of VOCs to emissions from furniture, flooring, building materials and wall coverings, characterized by acetone, formaldehyde, and acetaldehyde and high contributions of glyoxal, benzaldehyde and methyl isobutyl ketone. Oil paints and adhesives were found to contribute 12% of VOCs, characterized by methyl butyl ketone, acetone, 1,2-dichloromethane, methyl isobutyl ketone and benzyl chloride. Similarly, in a study by (Q. Liu et al., 2014) in 128 residences in Beijing, 2 out of 4 sources resolved by PMFv.3 belonged to the building materials/furniture group, although a comparatively limited number of chemical species were considered (three VOCs and four carbonyl compounds).

VOCs emissions from materials in new buildings were investigated using PCA/APCA on formaldehyde and VOC data from 107 Korean apartments at pre-occupancy stage (Shin & Jo, 2012). The predominant (30%) resolved source was represented by 16 VOCs was associated with flooring materials. The second factor (17%) traced by five VOCs corresponded with wood panel and vinyl floor covering emissions. A third factor (10%) with a high loading of 3 VOCs, whilst a fourth factor (9%) linked to 2 VOCs was related to various adhesives emissions.

In another large-scale study, (Campagnolo et al., 2017) applied PCA on VOCs and aldehydes measured in 140 modern office rooms in eight European countries. Among the resolved factors, were i) flooring emissions (traced by 2-butoxyethanol, 2-ethylhexanol and propanal), ii) wooden-based products (traced by  $\alpha$ -pinene), and iii) latex-based products (traced by acrolein and

benzaldehyde). The authors underlined the importance of information collected in questionnaires and checklists regarding the materials, equipment and activities in each building. This information is of major importance during the source identification process, especially for VOCs related to more than one group of sources (Lv et al., 2019; Shang et al., 2019). Finally, in the study of Yadav & Devi, 2022, 25 flame retardants (FRs) detected in dust from 22 indoor environments in India were used in PMF and PCA to indicate a significant source contribution (80%) from household items such as plastics, textiles, polyurethane foam, anti-foam agents, PVC, paint, and coatings.

Other receptor models (except for PMF and PCA) were applied to building and furniture material emissions in a limited number of studies. For example, Poisance et al., 2017 applied CMB on VOC concentrations. More precisely, SA was attempted in three newly built timber frame houses based on the material emission data obtained in a test chamber. Most of the emission profiles were dominated by aldehydes, with the exception of wood-based materials that were sources of both terpenes and hexanal, while polyurethane adhesive mastic was associated with high emissions of ethylbenzene and xylenes. The study indicated that the apportionment of VOC source contributions appears to be highly dependent on the position of source materials in the building (i.e. surface materials or internal materials) as well as on ventilation conditions. Poulhet et al., 2014 followed a measurement procedure to apportion formaldehyde emissions from building and furnishing materials in French public schools. More than 29 sources of formaldehyde were characterized in each investigated classroom, with higher emissions from building materials compared to furnishing materials. With the use of a mass balance model, several strong emitters made of wood products and foam were highlighted. The ceiling was identified as the main source of formaldehyde in most classrooms.

Lastly, anthropogenic sources of zinc (Zn) and chrome (Cr) inside Portuguese and Spanish classrooms, related to indoor material coatings, have been reported by Canha et al., 2014; Rivas et al., 2015. The EF calculated for PM collected inside Portuguese classrooms indicated a Zn source which can be related to the outdoor and traffic related source, but also to specific emissions from several Zn-containing products applied indoors in order to protect steel, walls, wood surfaces, doors and windows. Conversely, higher indoor Cr levels (Spanish classrooms) than outdoors were associated with abrasion of the metallic parts of chairs and tables (Rivas et al., 2015).

### 3.4.2 Indoor combustion-related sources

The present review revealed that smoking is the most frequently identified source belonging to this group (40% of the relative studies), followed by burning of wood (23%), charcoal (23%), kerosene, candles and incense (<5%). Wood, charcoal and kerosene burning are reported as combustion resulting from heating or cooking. On the other hand, candles and incense burning are usually related to wellbeing cultural or religious activities. Combustion-related sources are typically traced by carbonaceous fractions, PAHs, specific metals, alkanes, water-soluble organic compounds (WSOC), and levoglucosan. The majority of the SA studies have therefore focused on PM chemical composition. A limited number of studies have used VOCs, aldehydes (formaldehyde) or hopanes for combustion source identification (Bari et al., 2015; Clougherty et al., 2011; Huang et al., 2019; H. Pekey et al., 2013; van Drooge et al., 2018).

Smoking or environmental tobacco smoke (ETS) has been identified as a source by SA studies performed primarily on different PM fractions. PM components that trace ETS are usually carbonaceous fractions (OC, EC, BC), PAHs (benzo(k)fluoranthene, benzo(ghi)perylene) and specific metals (K, Cd, Ni, Mg, Cu, As). Cerium (Ce) revealed a smoking source profile in

Minguillón et al., 2012 and Pey et al., 2013, as it is attributed to the use of lighters. A number of SA studies using VOCs, ketones and aldehydes (Table 1) as key species have distinguished smoking as a separate indoor source (Huang et al., 2019; Masih et al., 2012; Pekey et al., 2010; Pekey et al., 2013; Yang et al., 2018), a source characterized as a mix of combustion processes (Bari et al., 2015), or combined with specific different sources i.e., soil dust (Barraza et al., 2014) and vehicles emissions (Othman et al., 2019). In some cases, activity recording by the occupants (e.g. frequency and duration of smoking) has assisted this source distinction (Barraza et al., 2014; D. E. Saraga et al., 2010; Shang et al., 2019).

The variability of the smoking source is another characteristic highlighted in relative SA studies. Tunno et al., 2016 demonstrated that tracers of the smoking factor resolved by PMFv.5 (i.e. black carbon, cadmium and potassium) experienced greater variability than the outdoor sources, even in heavily-industrialized communities. Cadmium was found to be the most unpredictable compound (Shang et al., 2019; Yang et al., 2018) with significant differences between populations at different sites, vocations and smoking exposures. In general, our review indicated that the source contribution of smoking ranged substantially from <10% (Minguillón et al., 2012) to more than 60% (Tunno et al., 2016) of indoor PM<sub>2.5</sub> (Figure 2a); however a much smaller range (10-22%) is noticed for VOCs (Figure 2b). Finally, it is also remarkable that ETS has been identified even in buildings with a smoking ban, as in the PCA study of Othman et al., 2019, where the source of smoking was attributed to ETS transfer by the smokers themselves (e.g. via clothes or skin).

Biomass combustion emissions (e.g. from fireplace or woodstove burning) vary between different locations due to the type of burning materials (D. E. Saraga et al., 2015). Fingerprints of wood burning are K<sup>+</sup>, Cl, EC, BC, VOC, WSOC levoglucosan, mannosan, galactosan, glucose,

mannitol, etc. (Table 1). Indoor SA studies have associated the biomass combustion source either with outdoor origin (i.e. combustion-related pollution in the vicinity of the building), or with the indoor use of a wood stove or fireplace during the measurements. The study of Zhu et al., 2012 belongs to the first case, where PMF revealed that biomass burning emissions originating from outdoors, contributed to both outdoor and indoor  $PM_{2.5}$  mass (44% and 31% respectively). In the second category, Canha et al., 2014 identified a significant contribution (60%, mixed with soil resuspension source) to indoor  $PM_{2.5}$  of wood burning (heating stoves) inside the classrooms of a rural school in Portugal. Similarly, Lai et al., 2019 used CMB model to apportion  $PM_{2.5}$  sources in 40 rural Chinese houses, where wood was the predominant residential fuel for heating. The selected biomass burning source profile was a composite of pine and mixed wood combustion in a Chinese residential wood stove, which eventually proved to have an overwhelming contribution to indoor PM. Firewood used for cooking in West Africa accounted for up to 87% of the total  $PM_{2.5}$  mass in cooking areas, as PMFv.3 revealed in Zhou et al., 2014. Biomass burning was represented by two distinct factors: fresh biomass smoke (characterized by K, Cl, S, and BC) and aged biomass particles (KCl and  $K_2SO_4$ ).

The impact of different fuel use for building heating on domestic respirable particulate matter (RPM) has also been investigated. Pervez et al., 2012 performed a two-step SA study in Central India, where they applied a linear regression analysis to evaluate the possible relationship between indoor-receptors and identified source routes and then CMB to quantify the relative source contributions. The contribution of household fuel burning emissions to the residential RPM varied between 17-48%, depending on the type of ventilation and fuel used in the houses (liquid petroleum, gas, kerosene, electric, coke and cow dung cake stoves).

A factor corresponding to residential coal burning (traced by high loadings of Pb, Cs and  $\text{Cl}^-$  and ranging between 5 and 20% of measured  $\text{PM}_{2.5}$ ), distinguished from large scale coal fired power plants emissions in China, was identified in the PMF study of Shang et al., 2019. Although coal has been replaced by gas for heating fuel in the urban area of Beijing since 2014, it is still used in some surrounding areas for cooking and space heating. In the study of Rybak et al., 2019, the SA of PAHs revealed the predominance of different types of combustion among the sources contributing to indoor concentrations. In particular, 15 PAHs were measured in spider webs in selected homes differing in location (rural area vs. city), type of room (living room, kitchen, basement), inhabitants' habits (smoking cigarettes vs. non-smoking) and type of heating/cooking devices used (natural gas, liquefied gas, coal- and wood-fueled heating). The application of cluster analysis (CA) suggested that the dominant sources were petrogenic and pyrogenic, originating from a mixture of sources, including cooking, smoking, heating and neighboring traffic activities.

Emissions from candle and incense burning have been associated with UFP or  $\text{PM}_{2.5}$  as well as PAHs,  $\text{NO}_x$ , nitrous acid ( $\text{HCNO}$ ), carbonyls and VOCs and are commonly associated with the cultural habits of occupants. Although these sources have been measured in both small-scale and large-scale chamber studies (Géhin et al., 2008; Glytsos et al., 2010), SA studies for their contribution to indoor pollution levels are relatively limited. Bekö et al., 2013 continuously measured ultrafine particle number concentration, PNC (10–300 nm in size), over a period of ~45 h in 56 residences of nonsmokers in Copenhagen, Denmark. Diary entries regarding occupancy and activities related to particle formation were used to identify source events and apportion the daily integrated exposure among sources. In homes where candle burning took place, more than half of the residential daily integrated exposure was attributable to this activity.

In another study, MacNeill et al., 2014 showed that candle burning increased the non-ambient component of indoor  $PM_{2.5}$  through the use of a computerized algorithm, which removed (censored) peaks due to indoor sources. The censored indoor/outdoor ratios were then used to estimate daily infiltration factors and to determine the ambient and non-ambient components of the total indoor concentrations. In both winter and summer, the majority of  $PM_{2.5}$  was of ambient origin (daily winter median = 59%; daily summer median = 84%). Masih et al., 2012 applied PCA on total PAH concentrations measured in urban Indian residences. The analysis revealed a source (30% of the variance) related with incense burning, though being combined with smoking source (traced by acenaphthene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(e)pyrene, benzo(a)pyrene and benzo(ghi)pyrene).

### 3.4.3 Cooking-related sources

Cooking has been characterized in SA studies either by specific activities i.e. frying, grilling, toasting Bekö et al., 2013; MacNeill et al., 2014; Men et al., 2021 or as an unspecified 'cooking source'. The overwhelming majority (>70%) of the SA studies identifying a cooking source, have been based on PM (mainly  $PM_{2.5}$ ) chemical composition. Only one study used UFP concentration, while five studies have used VOCs as the target pollutant (L. Huang et al., 2018; Y. Huang et al., 2019; Lundberg et al., 2021; MacNeill et al., 2014; Masih et al., 2012; Romagnoli et al., 2016; Won et al., 2021). In a literature review by Abdullahi et al., 2013, focusing on mass concentrations, size distribution and chemical composition of aerosols generated from typical styles of cooking, it was concluded that cooking can generate significant masses of aerosols belonging mainly to the  $PM_{2.5}$  and UFP range, and containing species such as alkanes, fatty acids, dicarboxylic acids, lactones, PAHs, alkanones and sterols (MacNeill et al., 2014).

Significant inconsistencies exist between indoor measurement data and published source profiles for cooking, which makes it difficult to obtain reliable estimates of the relative contribution of cooking to ambient concentrations (Robinson et al., 2006). Emissions from cooking are highly dependent on parameters including culinary techniques, cooking fuel, ingredients, cooking oil, relative humidity, cooking temperature, etc. (Eriksson et al., 2022). For instance, Abdullahi et al., 2013 showed that Chinese cooking can lead to a much greater contribution of PAHs in PM relative to Western-style fast food cooking. Western-style fast food cooking tracers were 9-octadecenoic, hexanedioic and nonanedioic acids, whilst Chinese cooking tracers were oleic acid, mannosan and galactosan. Emissions from heated stainless-steel cooking utensils (woks, spatulas), traced by Ni and Cr, were found to contribute to PM in 14 barbeque restaurants, as resolved by PCA and EF (Taner et al., 2013).

Cooking emissions have been investigated by several SA studies (Table 1). In their PCA study, Masih et al., 2012 identified a prevailing factor associated with cooking activity (36% of total PAHs variance, traced by benzo[ghi]perylene, benzo(b)fluoranthene, naphthalene, benzo(a)anthracene, chrysene, benzo(k)fluoranthene, benzo(e)pyrene). A separate source (31%, traced by methylnaphthalene, 1-methylnaphthalene, biphenyl, carbazole) was attributed to oil fumes inside houses in an Indian urban and roadside area. Specific trace elements have also been shown to trace cooking sources (Table 1). Othman et al., 2016 observed that high temperatures during cooking can promote the desorption of trace elements from cooking utensils and cooking oil, and release several trace elements from food ingredients, such as Fe and Cu from vegetables and meat. Yang et al., 2018 applied Varimax Rotated Factor Analysis on PM<sub>2.5</sub> collected from 47 residential Chinese buildings. Among the resolved sources, a cooking source was distinguished

(contributing 14%), by the abundance of Zn and Ca, which are elements included in cooking fumes.

An important observation with potential implications for SA studies was made by Kaltz et al., 2019 using Aerosol Mass Spectrometry (AMS) coupled with PMF in the HOMEChem project (The House Observations of Microbial and Environmental Chemistry). Using the default ionization response for organic species during cooking events, the AMS measured 4 to 10 times more aerosol mass than co-located instruments, which measured overlapping particle size distributions. The authors attributed the difference to a high ionization efficiency of long-chain fatty acids, such as oleic acid, which are primary components of cooking organic aerosols (COA). It was concluded that fresh COA can differ from ambient COA, which may go through physical and chemical aging upon strong dilution and oxidation outdoors, impacting the way it is measured in the AMS compared to fresh COA indoors.

The variation of the chemical composition of cooking emissions limits the application of receptor models (such as CMB), which need a priori knowledge of the source profiles. Arhami et al., 2010 applied CMB model to chemical components of indoor quasi-UFP (PAHs, hopanes, steranes) and highlighted the difficulty of apportioning the contribution of cooking, when using common meat cooking source profiles. Similarly, Lai et al., 2019 applied CMB on PM<sub>2.5</sub> samples from 40 households in rural China. Although the food cooking source profile chosen for their study was selected to be characteristic of the regional cooking style, mass not apportioned by the CMB model may still include some PM<sub>2.5</sub> from food cooking. Although factor analysis models (PMF, PCA) seem to overcome this difficulty, it is still challenging to identify specific cooking styles, as well as to achieve an optimal separation of the cooking organic aerosol factor from other contributing factors (Abdullahi et al., 2013).

### 3.4.4 Resuspension

Mineral dust can be introduced indoors by infiltration from outdoors, but also by being adhered to shoes and clothing, especially in schools with sandy playgrounds (Minguillón et al., 2015). Once there, mineral dust tends to be deposited, but can be frequently resuspended by convective flows. The dust resuspension source is usually identified by crustal elements in PM (Si, Fe, Al, Ca, Mg, Ti). In the majority of the cases, the resolved indoor resuspension factor is based on PM chemical characteristics similar to those of soil dust (Carrion-Maza et al., 2019; Hassan et al., 2020; Othman et al., 2016), indicating its outdoor origin and the re-emission of settled particles by indoor activities (Fromme et al., 2008). Our review indicated that the mean contribution of resuspension is clearly higher for PM<sub>10</sub> compared to PM<sub>2.5</sub> (Figure 2a).

In their PMF study, Molnár et al., 2014 concluded that although the elemental source profile for the indoor resuspension factor consisted of a combination of outdoor sources (long range transport, oil combustion, traffic, and soil), its day-to-day variation was indoor activity-driven and not governed by the fluctuation of outdoor concentrations. In line with this, Kopperud et al., 2004 observed that significant indoor activities such as cleaning and walking caused 60–90% of the daily indoor PM<sub>2.5</sub>. Even on low-activity days, when the house was unoccupied for most of the sampling period, 27–47% of the PM<sub>2.5</sub> originated indoors.

Resuspension of soil entering the indoor environment via footwear has been the subject of SA studies, especially in school or playground environments. For instance, Rivas et al., 2015 recorded very high PM<sub>2.5</sub> concentrations in schools with sandy playgrounds, with a high contribution of mineral dust elements, owing to the grounding down of mineral particles (usually in the coarse mode) after playground activities. In the same project (BREATHE), Amato et al.,

2014 applied PMF on indoor and outdoor PM<sub>2.5</sub> samples from 39 primary schools in Barcelona. They concluded that 47% of indoor PM<sub>2.5</sub> was generated indoors due to continuous resuspension of soil particles (13%) and various sources (34%) of organic (skin flakes, clothes fibers, possible condensation of VOCs) and Ca-rich particles (from chalk and building deterioration). It was shown that indoor mineral-source contributions were also affected by the orientation of the classroom relative to the unpaved playground. Where playgrounds were unpaved, the resolved mineral-source demonstrated a higher contribution indoors compared to the paved ones, particularly in street-adjacent classrooms.

In a recent PMF study, Carrion-Matta et al., 2019 identified two dust-related PM<sub>2.5</sub> sources inside 32 classrooms in the Northeastern U.S.: a Ca/Fe/Ti-rich source, possibly associated with outdoor road-dust or degradation of cement and 'dry-wall' indoors, and soil dust (traced by Al and Si). For both sources, the crustal PM can enter classrooms through windows or be released from the children's shoes and clothes, especially after playing outside. A similar finding was presented by (Mohamad et al., 2016) who applied PCA-APCS on PM<sub>10</sub> chemical composition measured in two schools (city centre vs suburban area) in Kuala Lumpur, Malaysia. Two indoor dust-resuspension factors were identified. One was a road-dust source traced by a strong loading of Cd and moderate loadings of Cu, Co, Pb, V and Mg<sup>2+</sup>. The second one, traced by Al, Ca<sup>2+</sup> and moderate loading of Zn and NH<sub>4</sub><sup>+</sup>, was associated with non-combustion motor vehicle emissions.

### **3.4.5 Cleaning and consumer products**

Cleaning and other consumer products have been identified in SA studies either as separate, specified sources or as a combined 'household products emissions' source (Table 1). Liquid household products (e.g., deodorizers, cleaners, colour removers, pesticides, and polishes) can

release several toxic aromatic and chlorinated organics, thus these sources are usually traced by VOCs (terpenes), carbonyls or halogenated organics (Halios et al., 2022). Ethanol is widely used as a common solvent in household cleaning agents such as bleach, dishwashing and laundry detergent, disinfectants, glass cleaners, fabric softeners, oven cleaners as well as deodorizers (Kwon et al., 2008; Singer et al., 2006). Formaldehyde and acetaldehyde can be released from cleaning reagents and floor cleaners (Salthammer et al., 2010). Acetaldehyde has been associated with air fresheners (Kwon et al., 2008; A. Steinemann, 2017). Acetone and ethyl acetate are emitted from nail polish remover, oil paint, furniture polish, wallpaper and carpet glues while 1-propanol can be found in dishwashing detergent (Heeley-Hill et al., 2021). 1,1-dichloroethane and methylene dichloride have been found in cleaning agents, pesticides and glues. Emissions from consumer products with fragrance, perfumes and essential oils (i.e., air fresheners, deodorizers, laundry detergents, fabric softeners, hand sanitizers, personal care products, soaps) are traced by terpenes such as limonene,  $\alpha$ , $\beta$ -pinene, styrene, p-cymene (A. Steinemann, 2017; Wallace et al., 1987; Yuan et al., 2010; Zuraimi et al., 2006). Our review indicated a similar mean value of four types of products (i.e. fresheners, cleaning products, paints, solvents) contribution (Figure 2b).

In the PMF study of Bar et al., 2015 performed on 117 VOCs collected inside 50 Canadian homes, three out of nine resolved factors corresponded to household and consumer product emissions. The dominant factor (44% of total measured VOCs) was interpreted as household product emissions and was traced by a large fraction (80%) of ethanol and moderate fractions of ethyl acetate, 1-propanol, acetaldehyde, acetone and 1,2-dichloroethane. Another factor (2%) corresponded to fragranced consumer product emissions and was distinguished by  $\alpha$ -pinene (73%),  $\beta$ -pinene (40%), camphene (46%) and small amounts of p-cymene, styrene and limonene.

The third factor (8%) was interpreted as deodorizers and was traced mainly by limonene,  $\beta$ -pinene, ethyl acetate, dodecane, and 1,4-dichlorobenzene (p-DCB). In another PMF study, Y. Huang et al., 2019 attributed 17% of total measured VOCs and carbonyls to a household products factor characterized by methylene chloride, acetone, acetaldehyde, chloroethane, 1,1-dichloroethane, cyclohexane and smaller amounts of BTEX. The PCA/APCS model used by Guo, 2011 on 16 VOC species identified in 100 Hong Kong homes revealed a room freshener (contribution 8%, traced by nonane, decane, 1,2,4-triethylbenzene and d-limonene) and other consumer products (6%, traced exclusively by methyl isobutyl ketone) as sources.

A cleaning-related source traced by a high proportion of Cl in PM<sub>2.5</sub> (>60%) has been identified by the PMF studies of Barraza et al., 2014 (households in Santiago, Chile) and Barmpareos et al., 2020 (classrooms in Athens, Greece). A source related to office floor cleaning characterized by significant fractions (>80%) of 2-butoxyethanol was revealed by the PCA study of Campagnolo et al., 2017, although the emissions from the flooring materials themselves were not excluded. The application of PCA on 25 VOCs measured inside a university classroom in China (Bai et al., 2022), revealed that four out of seven factors were associated with deodorants, cleaners and cosmetics, though it was not possible to distinctly identify each source (more than one factor was associated with the same tracer). However, the authors concluded that among VOCs detected in the classroom, PAHs originate from automobile exhaust emissions, while the main source of halogenated hydrocarbons and chlorinated hydrocarbons was the use of solvents.

Consumer products have been identified as emission sources in a few SA studies for PM. Apart from the aforementioned studies of Barraza et al., 2014 and Barmpareos et al., 2020, in the PMF analysis of Martins et al., 2016, emissions from aromatic musk products traced by methyl-dihydrojasmonate and galaxolide in PM<sub>2.5</sub> were identified inside four subway stations in

Barcelona. The study of Li et al., 2021 is the only one that identified cosmetics/personal care products as a source contributing to indoor residential dust, using phthalate esters (PAEs) as tracers. In particular, PCA and PMFv.5 were applied on eight PAEs detected in dust collected from 72 residences (bedrooms and living rooms) and 22 dormitories in China. Diethyl phthalate (DEP) and dimethyl phthalate (DMP) were the two specific PAEs that traced personal care products in both SA approaches, which resulted in different percentages of source contribution (PCA: 23%, PMF: 17.5% for houses; PCA: 22%, PMF: 37% for dormitories). Differences were also observed in the source chemical profiles between the different environments (residences vs. dormitories).

#### **3.4.6. Indoor-generated secondary pollutants**

Secondary pollutants generated indoors have been considered in several SA studies. The presence of VOCs together with the infiltration of oxidant radicals (such as OH, hydroxyl radical) and ozone ( $O_3$ ), might lead to the generation of oxygenated VOCs (OVOCs, such as formaldehyde), and secondary organic aerosols (Weschler & Carslaw, 2018, and references therein). These reactions can be driven by the emissions of VOCs from consumer products (Steinmann et al., 2011), building materials (Aoki & Tanabe, 2007), and cleaning products (Singer et al., 2006) in indoor environments.

Zhu et al., 2012 performed a  $PM_{2.5}$  SA study in nine Chinese residences. A PMF model found that the formation of secondary particles (including sulfate, nitrate and biomass aerosol) in indoor environments occurred from reactions of household products with  $O_3$  and hydroxyl radicals. They accounted for 23–43% of indoor  $PM_{2.5}$ . These formation processes led to a higher content of secondary organic aerosols (SOA) indoors than outdoors in summer, and approximately 1.5 times higher than in winter. Similarly, Arhami et al., 2010 observed that the

contribution of SOA during the warm season was about two times larger than during the colder season, highlighting the important role of photo-oxidation in the formation of SOA. The study revealed the sources of secondary particles after applying the CMB model on PAH, hopane and sterane concentrations measured in indoor quasi-UFP in elderly houses. SOA accounted for 3–19% of quasi-UFP mass, while being higher indoors than outdoors (up to approximately three times). This contribution was partially attributed to the formation of secondary particles in indoor environments from reactions of household products with  $O_3$  and to a lesser extent with OH radicals. Similar results were obtained by Reche et al., 2014 who reported that indoor UFP concentrations in school classrooms at midday were 30%–40% higher than outdoors, possibly due to SOA formation enhanced by indoor precursors or surface chemistry reactions mediated by  $O_3$ . They also found very high UFP in some cases during the afternoon when the schools were closed and cleaning products were used, generating UFP due to interaction with oxidants.

Another group of secondary pollutants are aldehydes (e.g., formaldehyde, acetaldehyde) originating from  $O_3$ - or OH-initiated reactions with many common and reactive VOCs that contain unsaturated carbon-carbon bonds (Salthammer et al., 2010). In a VOC apportionment study performed for 140 modern office rooms (Campagnolo et al., 2017), one of the sources (resolved by PCA) was positively correlated with formaldehyde and acetaldehyde but negatively correlated with  $O_3$ , implying the formation of  $O_3$ -initiated oxidation products. Although not being the dominant source in terms of explained variance, the factor representing secondary formation (e.g.,  $O_3$ -initiated reactions) accounted for 16% of the variance in summer.

The most common indoor sources of  $O_3$ -reactive compounds are carpets, cleaning products, printers and other office equipment (Cacho et al., 2013). Other indoor sources have been reported to include emissions from the occupants themselves, air fresheners, natural rubber

adhesives, soiled fabrics, soiled ventilation filters and personal care products (Y. Liu et al., 2021; J. Xu et al., 2023; X. Xu et al., 2022). A relationship between O<sub>3</sub>-initiated indoor chemistry and the increase of ultraviolet radiation has been observed in a number of SA studies. In particular, Campagnolo et al., 2017 observed a clear seasonal variation of this secondary products factor, as it was present only during summer (higher O<sub>3</sub> levels). A correlation between indoor carbonyl concentrations and O<sub>3</sub>-initiated secondary reactions during summer has been also reported by L. Huang et al., 2018 who performed a PMFv.5 study for VOCs in 27 residences in Beijing. Secondary pollutant production was attributed either to ground level O<sub>3</sub> or to other indoor precursors (i.e., various unsaturated VOCs) with higher concentrations in the warmer season.

The presence of WSOC has recently emerged as an important constituent of indoor air PM<sub>2.5</sub> (Duarte & Duarte, 2021; Duncan et al., 2019). The investigation of the composition of this organic aerosol component is much more developed for outdoor environments than indoors, mostly due to the key role of WSOC in climate and human health. In outdoor environments, it has been reported that WSOC mainly consist of a highly diverse suite of oxygenated compounds, including dicarboxylic acids, keto-carboxylic acids, aliphatic aldehydes and alcohols, saccharides, saccharide anhydrides, aromatic acids, phenols, but also amines, amino acids, organic nitrates, and organic sulfates, originating from both secondary processes and primary emission sources (e.g., biomass combustion, marine aerosols) (e.g., Duarte et al., 2021 and references therein). In indoor settings, on the other hand, the available knowledge on the WSOC component is still limited to its contribution to indoor particulate matter mass load. For example, Hasheminassab et al., 2014 reported that WSOC accounted for a highly variable fraction (1% to 13%) of PM<sub>2.5</sub> collected in three retirement communities of the Los Angeles Basin. In their CMB study, Lai et al., 2019, on the other hand, reported that WSOC were the most important

constituent of household  $PM_{2.5}$ , accounting for up to 70% of  $PM_{2.5}$  mass, with higher levels in winter than in summer. A similar seasonal trend has been also reported by W. Huang et al., 2015 in rural Chinese homes. Aerosol WSOC have also been considered as key components to be measured when dealing with PM-induced damages of works of art displayed in museums, particularly because they may contain chemically reactive species. In this regard, Daher et al., 2011 conducted a chemical characterization and SA study of  $PM_{2.5}$  inside the refectory of Santa Maria Delle Grazie Church, Home of Leonardo Da Vinci's "Last Supper", reporting that WSOC accounted on average for 18% of the  $PM_{2.5}$  mass.

The importance of aerosol WSOC for indoor SA studies assumes that this organic aerosol fraction is an indicator of SOA formation. Based on this assumption, and using a CMB model, Arhami et al., 2010 estimated that SOA contributed up to 19% of the measured mass of quasi-UFP ( $PM_{0.25}$ ) in indoor environments at four retirement communities in California. Nevertheless, the authors also highlighted that indoor SOA estimates based on the aerosol WSOC content should be determined with caution, since the latter can have contributions from primary indoor emissions of water-soluble organic particulate species (such as those from cooking). High concentrations of indoor WSOC and levoglucosan (water-soluble species and a key tracer of biomass burning) have been also used to identify (by PMF analysis) a wood combustion source in rural Chinese homes (W. Huang et al., 2015).

Hasheminassab et al., 2014 highlighted that indoor WSOC can be impacted by primary PM, such as those from biomass burning, cleaning and other consumer products that release water-soluble organic acids. In fact, levoglucosan is a primary PM component. An additional source of uncertainty in the estimate of SOA based on WSOC content is associated with the multiplication factor used to convert WSOC into WSO matter (WSOM) (organic matter-to-organic carbon ratio of 2.5) and with the assumed fraction of water-insoluble organic carbon in SOA (20%), as they both can vary with time and location (Arhami et al., 2010 and references therein). This aerosol water-soluble organic component is chemically dynamic and continuously evolving via oxidative chemistry. Several studies have explored this evolution and the chemical characteristics of WSOM in the outdoor atmosphere but none have investigated it indoors, suggesting that these topics should be addressed in future indoor studies.

### 3.4.7 Other sources

Source apportionment for organic matter (OM) in PM has been carried out in houses (Almeida et al., 2022; Tofful et al., 2021) schools (Almeida et al., 2022; Amato et al., 2014) and in other locations such as the refectory of Santa Maria Delle Grazie Church (Daher et al., 2011). Humans are considered to be one of the most important sources of OM in coarse PM, since they directly emit primary biological material (e.g., skin flakes, hair). According to the abovementioned studies, the total OM source contribution inside houses and schools ranged between 24% and 54% in  $PM_{2.5}$  and between 15% and 43% in  $PM_{10}$ , depending on anthropogenic factors such as occupant density and dust resuspension. In the case of the Santa Maria Delle Grazie Church, the CMB model run by (Daher et al., 2011) indicated that the OM source (dust of biogenic origin, with emissions from waxes used in the restoration process of the painting, or from human skin)

accounted for  $81 \pm 17 \%$  of the indoor  $PM_{2.5}$ , with  $78 \pm 3 \%$  being water-insoluble. It is noteworthy that the measured palmitoleic and oleic acids as well as squalene (associated with human skin oils, restoration waxes and skin care products) exhibited a similar seasonal trend as the OM source.

Xie et al., 2021 investigated whether quantitative fatty acid signature analysis (QFASA) of dust, could be applied to indicate sources and their respective contributions for a major class of indoor organic pollutants organophosphate flame retardants (OPFRs). In particular, significant correlations were observed between OPFR concentrations and lipid content in dust from Chinese and Australian houses. Using fifteen signature fatty acids in various indoor sources and the QFASA model, they found that clothing was the predominant contributing factor of dust OPFR (39% in Australia and 37% in China), followed by cooking oil (35% Australia, 26% China), pet hair (21% Australia, 35% China) and dandruff (4% Australia, 3% China)

Another source of indoor PM is fireworks. Camilleri et al., 2022 have isolated, through PMFv.5, a fireworks factor (contribution 6% to indoor  $PM_{2.5}$ ), which was traced by high percentages of Sr (60%) and Ba (35%). The authors also noticed that the meaningful contribution (37%) of K in this factor profile could possibly be attributed to the prevalence of large amounts of K salts in pyrotechnic compositions. This factor's contribution is characteristically seasonal (between June and September), matching the traditional local feast season. The same tracers (K, Ba, Sr, added to fireworks to create red-yellow colors), used in the PMF study of Matthaios et al., 2021, indicated a mixed source of fireworks and environmental tobacco smoke emissions contributing to indoor  $PM_{2.5}$  (by 7%), in a total of 340 homes in Massachusetts, USA. While lower peaks in the time variation of this factor were likely related to smoking activity and meat grilling, specific peaks were likely related to fireworks on New Year's Eve and Independence Day.

A source exclusively identified by particulate matter SA studies performed in classrooms is blackboard chalk. In all cases, the resolved source is combined with dust resuspension (Amato et al., 2014; Canha et al., 2014a,b; Mohamad et al., 2016) as a result of movement or cleaning activities. As chalk consists mainly of  $\text{Ca}_2\text{SO}_4$ , and  $\text{CaCO}_3$ , this source is characteristically traced by Ca or  $\text{Ca}^{2+}$  (usually strongly correlated with  $\text{SO}_4^{2-}$  during school hours) and in some cases Sr, which due to a similar atomic diameter tends to substitute a proportion of Ca in these minerals. The contribution of the mixed source was found to range between 19% and 60% of the coarse PM fraction.

### 3.4.8 The role of outdoor air in indoor SA studies

The outdoor environment is typically represented by several distinguished outdoor PM sources: vehicle emissions, combustion-related sources, industrial emissions, long-range transport, crustal sources/dust, secondary aerosol and sea salt. The contribution to the measured pollutants indoors varies considerably (<10% - 90% of PM). SA models identify outdoor sources either by using only indoor concentrations, by analyzing indoor and outdoor data separately or by combining indoor and outdoor input datasets in one array, which has been proved to give the most robust results (Amato et al., 2014; Minguillón et al., 2012).

Regarding PM, receptor models can adequately distinguish outdoor and indoor sources. Substantial total contributions of outdoor sources to indoor PM concentrations have been reported in 33 schools in Barcelona (Amato et al., 2014) (53%); in elderly care centres, Lisbon (Portugal) (Almeida-Silva et al., 2016) (58%); 47 households in Santiago de Chile (Barraza et al., 2014) (50%); residential houses in Beijing (China) (Yang et al., 2018) (54%); rural areas Northwestern China (Zhu et al., 2012) (55-69%); 51 homes in urban and suburban areas downtown Toronto (Canada) (Jeong et al., 2019) (77%); school classrooms in New Zealand

(Trompetter et al., 2018) (66%). However, there are cases that almost the 100% of the identified sources in indoor SA studies are outdoor sources e.g. in 32 inner-city schools in the Northeastern US (Carrion-Matta et al., 2019) (95%), or residences in Beijing (China) (Xie and Zhao, 2018) (100%). This may be attributed to the study design, the selection of species and SA approach, or the specific purpose of each study. Several mechanisms, such as infiltration, ventilation, and deposition followed by resuspension, may significantly affect the contributions to indoor concentrations.

Indoor PM levels predominantly driven by crustal sources (e.g., soil brought in from outside on clothes and footwear) and resuspension (i.e. occupants' movement and activities) are of special interest in SA studies, especially those in school environments (Amato et al., 2014; Carrion-Matta et al., 2019; Trompetter et al., 2018) (see Section 3.4.4 Resuspension). Traffic is another common outdoor source identified indoors. Amato et al., 2014 discussed the influence of school building orientation on the contribution of traffic to indoor PM<sub>2.5</sub>. Their PMF results indicated that the median traffic source contribution to outdoor receptors facing the street ( $6 \mu\text{g}/\text{m}^3$ ) was 50% higher than the median for those facing the interior of the block ( $4 \mu\text{g}/\text{m}^3$ ). This increase was even clearer inside the classroom, with a factor of 2 between classrooms facing a playground ( $3 \mu\text{g}/\text{m}^3$ ) and those facing the street ( $7 \mu\text{g}/\text{m}^3$ ). In the same campaign, Reche et al., 2014 emphasized the impact of the location of schools in heavily trafficked areas, causing an increase in the abundance of quasi-UFP, which can effectively infiltrate. The authors highlighted the importance of urban planning to reduce the health risks on children.

Cao et al., 2019 collected composite settled dust samples from four types of microenvironments (offices, hotels, dormitories and kindergartens) in Beijing, and studied the particle size distribution, their origin and spatial variation. Diagnostic ratios and PCA indicated the

substantial contribution of fuel combustion to PAHs in the indoor dust samples (biomass and coal combustion 58%, and traffic emission 12%). Indoor/outdoor diagnostic ratios and factor analysis for PAHs were also used to identify PAH sources inside schools in Kaunas (Krugly et al., 2014). Indicatively, the ratios of benzo(a)anthracene/chrysene and benzo(a)pyrene/benzo(ghi)perylene indicated fuel combustion and/or vehicular emissions, while fluoranthene/(fluoranthene + pyrene) and indeno(1,2,3-cd)pyrene/benzo(ghi)perylene ratios indicated wood and/or coal combustion. The authors highlighted the influence of the volatility and phase-transformation of individual PAHs on factor analysis, as significant differences were observed between indoor and outdoor samples.

Indoor-to-outdoor (I/O) ratios for the resolved PM source contribution (i.e. % contribution of source A indoors / % contribution of source A outdoors) have been used to assist the SA procedure. For instance, in the study conducted by Minguillón et al., 2012 in 54 homes in Barcelona, the median I/O ratio for the estimated source contributions from traffic sources was about 0.75, which is similar to that found by Hasheminassab et al., 2014 in retirement homes in USA (i.e. median:  $0.74 \pm 0.34$ ).

In a different approach applied on data from 43 homes in Boston, USA, Clougherty et al., 2011 regressed indoor against outdoor PM and NO<sub>2</sub> concentrations, modified by ventilation, isolating the indoor-attributable fraction, and then applied constrained FA to identify source factors for indoor concentrations. The authors developed Land Use Regression predictive models using GIS-based outdoor source indicators, as well as a questionnaire on indoor sources, which showed limited predictive power, but corroborated some indoor and outdoor factor interpretations.

The segregation of outdoor and indoor sources through receptor modeling for VOCs seems to be more complicated than that for PM. For instance, Campagnolo et al., 2017 reported that the

indoor or outdoor sources of BTEX and n-hexane cannot be adequately distinguished. Although they are associated mainly with vehicles and industry emissions, local indoor sources (e.g., paints, adhesives, varnishes, solvents) cannot be excluded. In another SA study targeting VOCs, Rösch et al., 2014 stated that although the traffic source was mixed with indoor sources, there was no clear evidence that it affected the indoor concentrations of toluene and benzene as a result of ventilation. Y. Huang et al., 2019 used the I/O value of certain VOCs such as toluene, benzene, methylene chloride, propylene, n-hexane, n-heptane, glyoxal, acrolein and freon-11 (highly correlated with vehicle emission, biomass burning, industrial emission and solvent usage), to derive an outdoor factor, which accounted for a very low percentage (2.1%) of indoor VOCs. On the contrary, the mass balance method applied by Jia et al., 2010 on 17 measured VOC concentrations from ten mixed-use buildings in southeast Michigan, USA, indicated that the contribution of outdoor sources to indoor VOCs (including carbon tetrachloride and benzene) reached  $84 \pm 29\%$ . Note that recent evidence has shown that outdoor VOC concentrations can be influenced by indoor VOC emissions. McDonald et al., 2018 estimated that about half of fossil fuel-related VOCs outdoors originate from sources mostly used indoors (consumer products). Hence, this high value might just indicate correlation, not really sources.

Considering these findings, the typical ambient outdoor sources (traffic-related sources, long-range transport, combustion sources, natural sources) contribute to the indoor air pollutant levels to various degrees, depending on local and regional conditions (e.g., vehicle fleet composition, building types and ventilation, climate, season, industrial activity nearby the sampling stations) as well as the selected SA technique. Additionally, the chemical profiles of pollutants entering from outdoors are likely to evolve due to physicochemical processes taking place indoors. To overcome this limitation, Almeida et al., 2022 performed a novel PMF analysis using the

Multilinear Engine-2 (ME-2) solver and the SoFi (Source Finder) 8 Pro tool, achieving an accurate discrimination of the outdoor and exclusively indoor PM sources. According to their methodology, the outdoor PM sources were firstly identified by performing fully unconstrained analysis on the collected datasets. Then, the indoor data SA analysis was performed by constraining the outdoor profiles on the indoor runs with an  $\alpha$ -value of 0.2 (20% change allowed), and adding an additional unconstrained source. By using this approach, the estimation of the influence of the outdoor sources in the indoor environment was optimized.

In another study, Cattaneo et al., 2016 used a combined, three-tier approach which identified the infiltration of outdoor PAHs into indoor environments as the most important source in winter, with a relevant role played by biomass burning and traffic exhaust in both seasons. In particular, diagnostic ratios with Principal Component and Hierarchical Cluster Analyses (PCA and HCA), CMB and linear mixed models (LMMs) were applied on 15 PAHs identified in  $PM_{2.5}$  in 19 residences in northern Italy. The SA analysis was combined with multivariate statistics using building characteristics, HVAC systems and behavioral information collected by checklists and time activity diaries, for a more complete identification of PAH sources and determinants in the residential environment.

A more accurate estimation of outdoor contribution to indoor dust has been presented by Rasmussen et al., 2022. The use of volume-normalized element concentrations ( $ng/m^3$ ) in SA for evaluating the indoor/outdoor relationships resulted in an underestimation of indoor sources. However, by using mass-normalized metrics ( $mg/kg$ ), new evidence for indoor sources of several elements (such as S and Zn) was revealed (e.g. consumer spray products), which were previously assumed to be conservative tracers of outdoor particles.

Recent studies have focused on the impact of physicochemical processes occurring as pollutants are transported from outdoors to indoors. In Garbarienė et al., 2022, a direct comparative analysis of chemical composition and SA for indoor and outdoor PM<sub>1</sub> was performed in a building with a high-efficiency filtration system. Enrichment factors and I/O ratios revealed that resuspension and combustion sources prevailed outdoors, but due to significant filtration losses, pollutant infiltration was low. The altered chemical composition of indoor PM<sub>1</sub> was attributed to species-specific evaporation (depended on outdoor to indoor temperature gradient and relative humidity gradient) and some minor indoor sources.

In another recent SA study, Stratigou et al., 2022 used concomitant indoor (unoccupied building) and outdoor time-resolved measurements of inorganic trace gases, VOCs and PM<sub>1</sub> composition using online mass spectrometers to apportion indoor and outdoor contributions. The study identified whether the indoor environment acted as a source or a sink of these species, and it is one of the first that discusses the physicochemical processes affecting the composition of infiltrated air and the SA methodology. The results showed that this unoccupied indoor environment acts as a source of VOCs contributing 87% to OVOCs levels and 6% to C<sub>x</sub>H<sub>y</sub>, while it acts as a sink for O<sub>3</sub> (owing to high reactivity with alkenes present in ambient air or adsorbed on surfaces) and PM (likely due to up to 60% losses through volatilization).

#### **4. Challenges for indoor air pollution source apportionment**

As reported by (Hopke, 2016), the SA of ambient (outdoor) air pollution is now relatively mature in terms of its methods and its ability to adapt to new measurement technologies, so a high likelihood of extracting the maximal information from the collected data is assured. However, this is not the case for the SA of indoor air pollution. Our literature review revealed a number of

research gaps, which will challenge future studies on indoor air pollution SA and are now discussed in detail.

#### **4.1. Optimization of indoor air quality monitoring and data selection**

There are a number of limitations on IAQ monitoring that can indirectly affect receptor modeling performance: integrated indoor air sampling is labor-intensive and costly; PM gravimetric sampling indoors can result in low PM mass loading and consequently limited ability to measure chemical components; indoor air samples are usually time-averaged, not offering long-term time series and not capturing peak events; data collected from one sampling site are not representative of a whole house or building.

New developments in IAQ monitoring, i.e. online and/or real-time instrumentation (PM composition or size distribution, organic compounds) and sensor networks, can help to at least partially overcome these limitations (Andenas Garcia et al., 2022; Bergmans et al., 2022). Indeed, the latest SA studies underline the importance of capturing the temporal variability of the sources. However, even very high time resolution over a few days may provide low representativeness, given the very important seasonal variability of the sources, ventilation, infiltration and reaction processes.

The need for multiple indoor sampling/monitoring sites is also highlighted. The use of combined multiple site data could lead to improved source resolution and reduced rotational ambiguity, as achieved for outdoor SA (Kara et al., 2015). Thus, real-time monitoring over longer periods and/or data of higher spatial (i.e. monitoring networks) and temporal resolution provided by modern monitoring techniques already applied for outdoor air [e.g. online instruments for in situ PM chemical analysis as energy dispersive X-ray fluorescence (XRF), OC/EC and ion

chromatographic analyzers, or aerosol chemical speciation monitors (ACSM)] can increase modeling optimization. However, data from low or middle cost sensors located indoors are currently used with caution in SA studies, although such IAQ monitoring offers the advantage of a large amount of data (Bergmans et al., 2022).

Another crucial point is the specification of the chemical signatures of indoor sources, even though their a priori knowledge is not necessary for some widely used receptor models (e.g. PMF, PCA). While a respectable number of outdoor source profiles have been adequately measured worldwide (i.e. Bi et al., 2019; Pernigotti et al., 2016; <https://www.epa.gov/air-emissions-modeling/speciate>), corresponding information for indoor sources is limited. The present review revealed the need to specify the chemical profiles of more indoor sources as the different cooking styles and emissions from specific types of materials and consumer products. A detailed recording of specific indoor source profiles in each microenvironment is practically impossible, but further information on the emission type and strength would assist the source discrimination process. This is especially the case for VOCs, aldehydes and carbonyl compounds, which can originate from a wide variety of sources. Even if SA studies yield qualitatively similar results reflecting commonalities in indoor VOC sources, improved source signature data would improve quantitative comparison across different SA analyses.

Another challenge for future SA studies is to optimize the estimation of source contributions for indoor UFPs, given that the main metric for this fraction is the rapidly changing particle number concentration. New monitoring techniques can assist in this direction (Bergmans et al., 2022); however, targeted mathematical approaches are needed to examine particle number time series and identify potential contributing sources to episodic particle number concentration peaks.

## 4.2. Incorporation of physical and chemical processes in source apportionment methodology

Indoor air pollutant concentrations and characteristics are influenced by complex physical and chemical processes (e.g., partitioning, chemical transformations in the gas phase or on surfaces, etc.). For example, ammonium nitrate (a relevant component of outdoor  $PM_{2.5}$ ) can be volatilized indoors to yield  $NH_3$  and  $HNO_3$ , which in turn can react to yield other PM components (Lunden et al., 2003; Y. Xie & Zhao, 2018). In the effort to distinguish and quantify these emissions, SA models often make a number of assumptions, such as: source profiles are stable over time; significantly correlated compounds originate from the same source; sets of observations exhibiting a large temporal variability are able to deconvolve the different sources.

Therefore, future SA studies should incorporate the dynamics of source profiles. For example, as the evaporative loss of volatile or semi-volatile species depends on temperature and humidity gradients, there is a need to apply SA under different environmental conditions, but also in different seasons and for varying ventilation and infiltration conditions. Regarding linearity, mechanisms like infiltration, inertial deposition, diffusive removal processes and indoor mixing cannot be represented by linear relationships. Thus, a remaining research question is how these processes can be considered in a hybridization of physical models with factorial source separation methods. Moreover, information on time-activity patterns should be incorporated in SA procedures. Although the most often used receptor models adopt the realistic non-negative constraint on factor profiles and contributions, specific occupant activities (i.e. windows opening to increase ventilation, or interventions affecting deposition/adsorption processes) can impact the initially assumed conditions.

Recent trends in SA tools -applied to ambient air data- have improved source specification. To begin with, multidimensional data arrays are being used in specific factor analysis models i.e. Non-negative Tensor Factorization, which could also offer valuable new insights for IAQ modeling e.g. by combining IAQ and occupants activity or ventilation data. Moreover, new directions in factorization modeling aim to a better modeling of the temporal evolution of sources by using advanced computational techniques on long term ambient air quality data. For example, Bhandari et al., 2022 applied a *time-of-day* PMF on long-term organic aerosol datasets, to capture diurnal time series of patterns of sources by separating each day into smaller periods with low variability in emissions and meteorological conditions. In the same direction, a new methodology, named *seasonal* PMF, recommends PMF application on long-term ACSM organic aerosol datasets under the Source Finder (SoFi) Pro software package (Datalystica Ltd.) and identifies the major groups of organic compounds, providing seasonal but not an intra-seasonal variation of factor profiles (Sun et al., 2018). The more recently developed *rolling* PMF (Canonaco et al., 2021) applies the model on moving/rolling windows of a selected length over the entire dataset, and therefore it accounts for the temporal evolution of the source fingerprints. Recent studies indicate that *rolling* PMF is more accurate than *seasonal* PMF due to its profile-adaptation feature and its lower computational and evaluation time (Via et al., 2022).

Finally, there are now several studies using Bayesian approaches which assume that source contributions and profiles vary probabilistically (Park & Lee, 2019). As Hopke et al., 2020 underline, there may be some information about the profiles similar to what is used in CMB analyses, but very little is known about the distributions underlying the measured profile values since there have not been sufficient measurements made on any source at any time.

In conclusion, the abovementioned developments can substantially benefit indoor air SA methodology, although it is still unclear how they can overcome the limitations previously discussed.

## 5. Conclusions

This work presents a review of the recent scientific literature on source apportionment studies for indoor air. 127 papers were selected for further investigation on the specific SA methods used, the targeted pollutants and the resolved sources, as well as the limitations and knowledge gaps of this research field. The main conclusions from the present study are:

- Almost half of the indoor SA studies were conducted in residential environments, followed by schools/university and office buildings.
- PMF, PCA and CMB were the most frequently used receptor models.
- The overwhelming majority of the studies (78%) used PM or dust chemical composition or PM size distribution as tracers to apportion the measured concentrations to their potential sources. The remaining 22% performed SA for organic compounds.
- The source categories revealed by the indoor SA literature are as follows: i) building materials and furniture, ii) indoor combustions, iii) cooking, iv) resuspension, v) cleaning and consumer products, vi) secondary pollutants formation, and vii) other products and activities.
- In contrast to PM, organic compounds (VOCs, carbonyls, aldehydes etc.) can originate from a wider range of indoor sources. The characteristics of these sources depend on several factors, a fact that complicates the SA procedure. Most of the studies underline the need for more comprehensive research on the characterization of sources, e.g., by

specifying the detailed chemical profiles of sources with highly varying characteristics (different cooking styles, consumer products, etc.)

- The contribution of the outdoor environment to the air pollutant concentrations indoors varies considerably among the studies. The typical outdoor sources (traffic-related sources, long-range transport, combustion sources, natural sources) contribute to a differing extent to the indoor levels. Their contribution may differ due to local and regional conditions, as well as the selected SA technique. Additionally, chemical profiles of pollutants from outdoor sources evolve as they enter the indoor environment, due to physicochemical processes taking place indoors. Conversely, the influence of indoor VOC emissions is being increasingly recognized as an important contributor to outdoor VOC concentrations (McDonald et al., 2013).
- A number of research gaps regarding indoor air pollution SA were highlighted, including optimization of IAQ monitoring and data selection and the incorporation of indoor air physical and chemical processes in the already developed SA methodology.

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**CRedit author statement**

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**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests, personal relationships which may be considered as potential competing interests:

## Graphical abstract



Source group	Contribution to PM2.5
building materials & furniture	5-25%
combustion-related	2-61%
cooking	3-38%
resuspension	2-35%
consumer products	3-37%
indoor generated secondary pollutants	<10-45%
outdoor sources	10-90%

**Highlights:**

- the identified sources indoors, their tracers and the SA method used are summarized
- the number/type of identified sources strongly depends on the tracers used for SA
- 78% of the indoor SA studies focus on PM chemical composition/size distribution
- SA for organic compounds is complicated due to the wide range of their sources

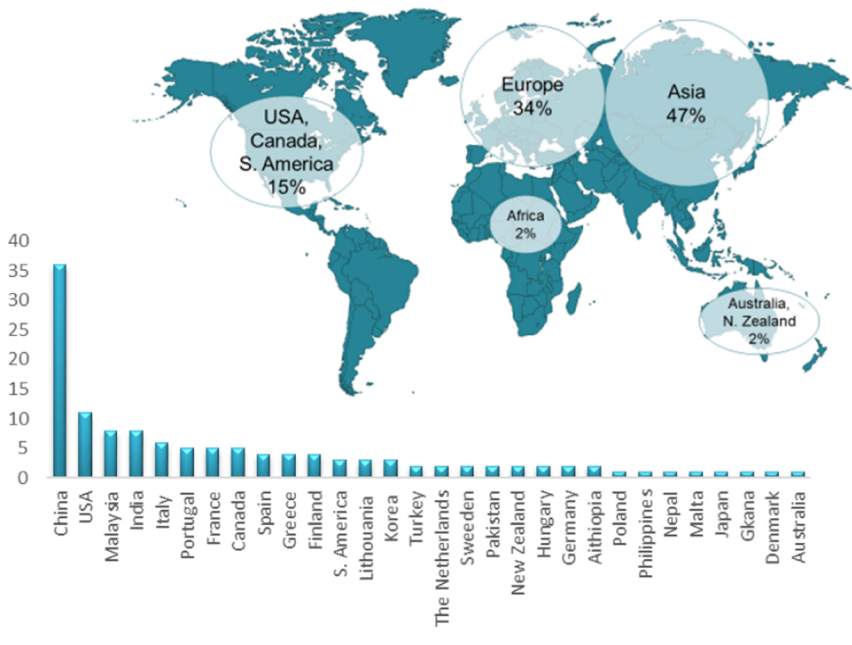
Journal Pre-proof



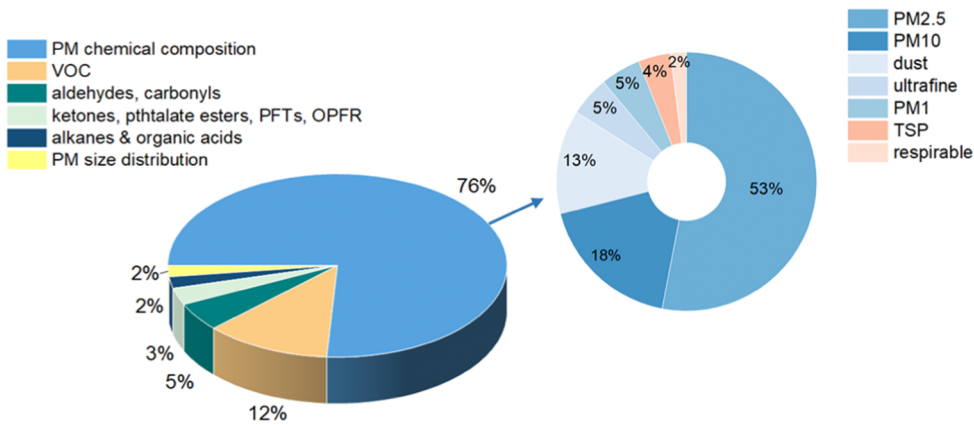
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Graphics Abstract

a.



b.



c.

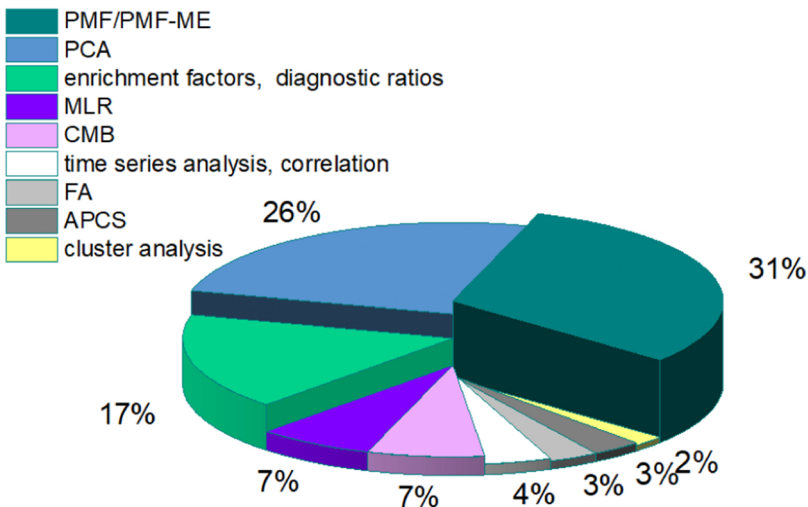
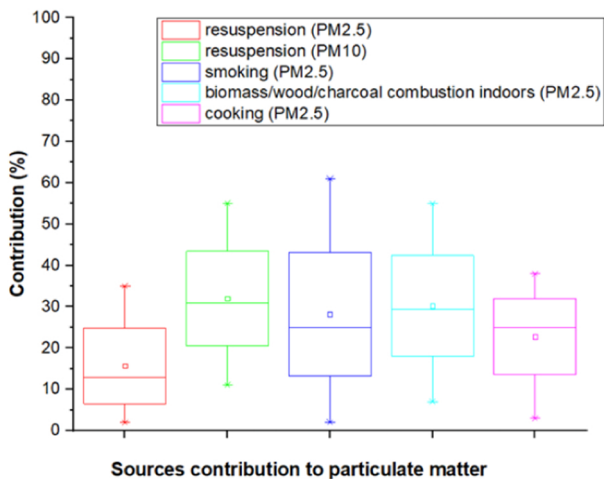


Figure 1

a)



b)

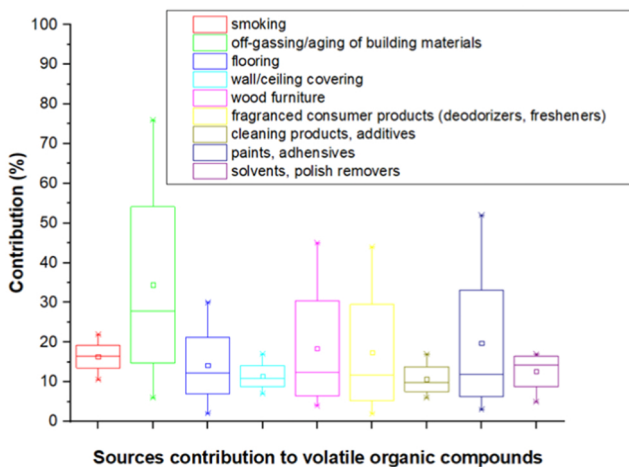


Figure 2