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Trends in indoor–outdoor $PM_{2.5}$ research: A systematic review of studies conducted during the last decade (2003–2013)

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ABSTRACT

There has been growing concern about potential health risks from exposure to $PM_{2.5}$ (fine particulate matter). The importance of conducting simultaneous indoor and outdoor measurements emerged because people, especially in developed countries, spend more than 90% of their time indoors. Great spatial and temporal variations in human exposure to $PM_{2.5}$ have recently been reported. This review aims to identify the main research areas that have attracted recent attention, any possible gaps in the measurements of $PM_{2.5}$ in various microenvironments, and the relationships between indoor and outdoor concentrations. This study also provides recommendations for further studies on $PM_{2.5}$ measurement methods and exposure levels. To achieve these goals, this review included articles published online from 2003 to 2013 in the ScienceDirect and Web of Science databases. In the initial screening stage, 113 abstracts selected while 61 articles were remained for full review. The reviewed studies consistently showed positive correlations between indoor and outdoor $PM_{2.5}$. Sulfate/sulfur concentrations were used intensively for calculating the infiltration factor (F_{INF}). The higher F_{INF} indicated high infiltration of outdoor $PM_{2.5}$ into indoor areas. Great percentage (42%) of the reviewed filter–based studies was conducted in Europe, followed by a similar amount (38%) in the USA, and 20% in Asia, indicating a lack in $PM_{2.5}$ research in other parts of the world. It was difficult to conclude that ambient fixed–site monitoring provided accurate estimations of actual exposure to $PM_{2.5}$. Studies shown trends of higher personal concentrations compared to indoor and outdoor ones. Higher indoor levels of OC (organic carbon), compared to outdoor levels, were consistently reported. The opposite trend was true for EC (elemental carbon), and there were higher indoor OC/EC ratios than outdoor OC/EC ratios. There was a consistent general trend of a high (r>0.70) correlation between indoor and outdoor EC, while the correlation between indoor and outdoor OC was much weaker (r=022–0.75). The higher indoor OC/EC ratios, compared to the outdoor OC/EC ratios, reflects multiple sources of indoor OC. Sulfate (SO_4^{2-}), nitrate (NO_3^-), and ammonium (NH_4^+) were primary contributors to $PM_{2.5}$ mass.

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1. Introduction

There is strong evidence for the potential and actual harmful effects of exposure to fine particulate matter ($PM_{2.5}$). $PM_{2.5}$ is a class of solid and liquid particles with an aerodynamic diameter ≤2.5 µm (Crist et al., 2008; Castro et al., 2010) which can pass through the respiratory system into deep parts of the lungs (Castro et al., 2010). Coarse particulate matter (PM_{10}) are particles with aerodynamic diameters <10 µm (Gadkari, 2010). The level of exposure to $PM_{2.5}$ is determined by a variety of factors including the size, shape, and chemical composition of the particle, personal characteristics (age, gender, socioeconomic status, nutritional status, and predisposing factors), weather conditions, and other factors of low impact (Wallace, 1996; Pope et al., 2004; Riediker et al., 2004; Gutierrez–Castillo et al., 2006; Baccarelli et al., 2008; He et al., 2010; Gualtieri et al., 2011; Myatt et al., 2011; Osornio–Vargas et al., 2011; Wang et al., 2013; Cachon et al., 2014).

It is well known that the composition of $PM_{2.5}$, particularly in urban areas, is quite complex because several different industrial, commercial, and residential sources contribute, in an unsystematic manner to the total $PM_{2.5}$ mass and its associated chemical composition (Monn, 2001). Accordingly, all possible pollution sources need to be assessed for both PM mass concentration and the associated chemical contents that can be used as source identifiers. Researchers commonly use different modeling approaches for source apportionment. The commonly identified major sources of ambient $PM_{2.5}$ in cities include secondary sulfate (SO_4^{2-}) and secondary nitrate (NO_3^-) (secondary aerosols), coal combustion, biomass burning, industrial emissions, motor vehicles exhaust, and road dust (Almeida et al., 2005; Song et al., 2006; Mooibroek et al., 2011; Jorquera and Barraza, 2012; Minguillon et al., 2012; Choi et al., 2013). Minor sources include sea salt, soil dust (Mooibroek et al., 2011; Jorquera and Barraza, 2012; Choi et al., 2013), and domestic heating (Kertesz et al., 2010).

The importance of conducting simultaneous indoor and outdoor measurements emerged for a variety of reasons: (1) most people, especially in developed countries, spend more than 90% of their time indoors (Bronsema et al., 2004; Cao et al., 2005; Alves et al., 2013); (2) gathering information from ambient monitors alone will either underestimate (Adgate et al., 2007) or overestimate (Cortez–Lugo et al., 2008) the actual exposure to $PM_{2.5}$; and (3) large spatial and temporal variations in exposure have been reported along with a relationship between indoor and outdoor air within certain microenvironments (Cyrys et al., 2004; Cao et al., 2005; Huang et al., 2007; Martuzevicius et al., 2008; Lim et al., 2011; Seleventi et al., 2012). Ambient air quality standards for particulate matter provide specific limits that should not be exceeded for outdoor levels of aerosols. However, for effective

management of particulate air pollution, such standards need to be integrated with national environmental policies because ambient levels of aerosols have exceeded the air quality standards in many countries (Gadkari, 2010; Pekey et al., 2010; Cao et al., 2012; Massey et al., 2012).

Involving community members in studies of personal and indoor measurements may not be as easy as conducting ambient monitoring of $PM_{2.5}$. This may be one reason why researchers adopted a wide variety of approaches for their measurements, which frequently provided incomparable results. Different studies have focused on different aspects of $PM_{2.5}$ exposure. The principal objectives of the current review are to identify the main research areas that have recently attracted the attention of researchers, identify the possible gaps in the research, and provide recommendations for further studies on $PM_{2.5}$ measurement methods and exposure levels.

2. Methodology

2.1. Reference guidance

The current study scrutinized the selected studies following guidelines for systematic reviews mentioned in detail in Torgerson (2003), Petticrew and Roberts (2006), Liberati et al. (2009), and Sumpter and Chandramohan (2013). First, the objectives of the study were identified. Second, potentially relevant studies were defined and screened for retrieval using the key words and synonyms mentioned in the following section. Third, the abstracts of the studies were reviewed in a group discussion to determine those that should remain for full review. Fourth, a subsequent assessment of these studies according to the inclusion criteria (described below) was conducted. Finally, there was a determination of the limitations of the review.

2.2. Search strategy

To retrieve the most relevant studies for inclusion in the review, the study question was broken into its three components. The search was then conducted using the key search terms of fine particulate matter along with the associated synonyms of fine particles, respirable particles, fine particulates, fine aerosols, and $PM_{2.5}$. Synonyms used for indoor and outdoor were I/O concentration, ambient, indoor, infiltration of ambient air into indoor, and indoor/outdoor. Search for [Title] was selected to capture all possible related articles using AND and AND/OR options for advanced searches.

2.3. Inclusion and exclusion criteria

The inclusion criteria were as follows: studies on fine particulate matter published online in the ScienceDirect and Web of Science databases. Only articles written in English and published from 2003–2013 were included. Some publications cited in the introduction and methodology sections were retrieved randomly to provide general background information about $PM_{2.5}$ or serve as guidelines for conducting the review, respectively.

To avoid duplication of publications, articles were first included based on their titles and abstracts. A total of 113 abstracts were retrieved in the initial screening stage using the key search terms mentioned in the "search strategy" above. After a careful reading of the selected articles, 61 were included. Duplicate articles were excluded by inserting all the selected publications into Endnote Software. Publications on PM in general and review papers were excluded.

3. General Overview of Indoor versus Outdoor Concentrations of $PM_{2.5}$

The vast majority of published studies on indoor–outdoor concentrations of $PM_{2.5}$ and their associated chemical composition showed positive relationships between the indoor and outdoor mass concentrations of $PM_{2.5}$ (Cyrys et al., 2004; Cao et al., 2005; Hovorka et al., 2005; Li et al., 2005; Ott and Siegmann, 2006; Huang et al., 2007; Martuzevicius et al., 2008; Lim et al., 2011; Massey et al., 2012; Seleventi et al., 2012). These correlations varied between moderately strong (r=0.52, p<0.05) in Sureda et al. (2012), strong (r=0.76, p<0.0001, overall median I/O ratio=0.79) in Cyrys et al. (2004) and (r=0.81, average I/O $PM_{2.5}$ ratio=1.4) in Cao et al. (2005), very strong (r=0.89, p<0.001, I/O $PM_{2.5}$ ratio=1.37) in Lim et al. (2011), and highly correlated (R^2=0.94) in Seleventi et al. (2012) when the major indoor sources (cleaning activities, cooking, and smoking) are omitted and varied between the seasons of the year elsewhere (Martuzevicius et al., 2008; Cao et al., 2012).

In general, an accurate measurement of particulate matter is challenging when different approaches of measurement give incomparable results because of the complex structure and nature of PM. The physical and chemical properties of the particles affect the mechanism of sampling in that the real–time samplers rely on light scattering while the active samplers depend on impaction of articles; both measuring techniques are influenced by the particles. Great contributions of outdoor $PM_{2.5}$ into indoor areas were recently observed (Hovorka et al., 2005; Li et al., 2005; Huang et al., 2007; Lim et al., 2011; Cao et al., 2012; Minguillon et al., 2012; Seleventi et al., 2012). Higher indoor concentrations, compared to outdoor concentrations of $PM_{2.5}$ have been reported and attributed to several sources including smoking (Sawant et al., 2004; Massey et al., 2012; Seleventi et al., 2012; Sureda et al., 2012), infiltration of vehicle exhaust or ambient air into indoor areas (Hanninen et al., 2004; Adgate et al., 2007; Baxter et al., 2007; Castro et al., 2010; Lim et al., 2011; Massey et al., 2012), cooking (Baxter et al., 2007; Gerharz et al., 2009; Brown et al., 2012), indoor heating during the winter (Jedrychowski et al., 2006; Massey et al., 2012), soil/crust–related activities (Larson et al., 2004; Lim et al., 2011), oil/coal combustion (Lim et al., 2011), road dust (Lim et al., 2011), miscellaneous indoor sources (Jedrychowski et al., 2006; Baxter et al., 2007), building materials (Hanninen et al., 2004), and cleaner sprays (Martuzevicius et al., 2008) (Figure 1). In contrast, low indoor levels of $PM_{2.5}$ could be explained by low indoor human activity (Cao et al., 2012; Sangiorgi et al., 2013). Table S1 (see the Supporting Material, SM) displays particles of indoor–outdoor $PM_{2.5}$ that depend completely or partially on active sampling using filtration methods.

Figure 1. *Summary of sources of indoor $PM_{2.5}$ ordered according to their potential contribution starting from the bottom of the hierarchy as the most significant contributors.*

4. $PM_{2.5}$ Chemical Composition

Based on the reviewed articles, Elemental carbon (EC), Organic carbon (OC), $PM_{2.5}$–bound PAHs (Polycyclic Aromatic Hydrocarbons), nitrate (NO_3^-), sulfate (SO_4^{2-}), and several trace elements have frequently been studied along with $PM_{2.5}$ mass concentrations. Indoor/outdoor (I/O) ratios of EC and OC varied considerably among seasons. Average I/O ratios of OC and EC were found to be 1.02 and 0.96 in Cao et al. (2012), 1.26 and 0.65 in

Seleventi et al. (2012), 1.02 and 0.8 in Ho et al. (2004), and 3.31 and 1.3 in Olson et al. (2008), respectively. This indicates that indoor OC was usually higher than outdoor OC and the opposite was true of EC, except in Olson et al. (2008) (Figure 2a). These trends were further clarified when average indoor OC/EC and average outdoor OC/EC were calculated and illustrated (Figure 2b) (Ho et al., 2004; Cao et al., 2005; Cao et al., 2012; Seleventi et al., 2012). These trends coincide with findings by Alves et al. (2013) where the OC content of $PM_{2.5}$ was found to be 4–5 times higher than EC, and partially agree with findings by Ho et al. (2004). OC was found to be the most abundant component of indoor $PM_{2.5}$ mass levels (Sawant et al., 2004; Alves et al., 2013). Ninety percent of the OC content was related to outdoor sources (Cao et al. 2012; Ho et al. 2004). Stronger correlations (correlation coefficient >0.70) between indoor and outdoor EC were reported than correlations between OC (correlation coefficient ranged between 0.22–0.75). These findings coincided with higher indoor OC/EC, compared to outdoor OC/EC, reflecting the existence of multiple sources of indoor OC (Cao et al., 2005; Olson et al., 2008; Zhu et al., 2012; Alves et al., 2013) (Figure 2).

Tobacco smoke, traffic (Castro et al., 2010), and wood smoke (Ott and Siegmann, 2006) were suggested as important sources of indoor PAHs, while vehicular emissions were a major outdoor source (Li et al., 2005). The low indoor/outdoor $PM_{2.5}$–bound PAHs ratio shown in Kliucininkas et al. (2011) indicates that these PAHs are from outdoor sources rather than indoor sources which is partially consistent with a study by Sangiorgi et al. (2013). Similarly, the correlation between indoor and outdoor PAHs suggests a contribution of outdoor sources to indoor levels and indicates widespread sources of PAHs (Li et al., 2005; Minguillon et al., 2012). In contrast, Olson et al. (2008) found that microenvironment variation accounted for 59% of observed PAHs variation, and PAHs with higher molecular weight had the highest average levels, possibly due to gasoline vehicle exhaust. Of 18 PAHs analyzed, benzo[*g,h,i*]perylene (BghiP) could be used as an indicator of PAHs air pollution in urban areas with high traffic (Li et al., 2005).

Water–soluble ions were substantially influenced by outdoor sources at schools (Alves et al., 2013), offices (Sangiorgi et al., 2013), and residential buildings (Meng et al., 2009; Seleventi et al., 2012). In all these microenvironments, calcium (Ca^{2+}) showed exceptional low I/O ratios indicating that, it was mainly of indoor origin. However, Huang et al. (2012) observed higher levels of these ions within office buildings and student dormitories, as compared to outdoor levels. Ions such as sulfate, nitrate, ammonium (NH_4^+), potassium (K^+), sodium (Na^+), magnesium (Mg^{2+}), and Ca^{2+} accounted for approximately a quarter (28%) and a half (49%) of the total mass of $PM_{2.5}$ in indoor and outdoor samples, respectively. Of these ions, SO_4^{2-} (Fromme et al., 2008; Seleventi et al., 2012; Zhu et al., 2012; Sangiorgi et al., 2013), NO_3^- (Zhu et al., 2012; Sangiorgi et al., 2013; Hassanvand et al., 2014), and NH_4^+ (Sangiorgi et al., 2013; Hassanvand et al., 2014) were found to have greater contributions than the other ions to both indoor and outdoor $PM_{2.5}$ mass concentrations. Calcium was one of the major contributors to indoor $PM_{2.5}$ (Fromme et al., 2008) even though both Ca^{2+} and K^+ are usually related to indoor sources (Long and Sarnat, 2004; Molnar et al., 2007; Meng et al., 2009; Zhu et al., 2012). Levels of the three secondary aerosols (NO_3^-, SO_4^2, and NH_4^+) decreased with increasing PM size (PM_1, $PM_{2.5}$, PM_{10}) (i.e., higher in $PM_{2.5}$ than in PM_{10}) (Hassanvand et al., 2014).

Levels of the elemental components of $PM_{2.5}$ in indoor–outdoor air in proximity to industrial areas and high–traffic roadsides were higher than those in ambient background environments (Huang et al., 2007; Gadkari, 2010). Based on enrichment factor analysis, some elements, manganese (Mn), aluminum (Al), iron (Fe), silicon (Si), and calcium (Ca) are considered crust–related whereas others [sulfur (S), lead (Pb), chlorine (Cl), and zinc (Zn)], are related to anthropogenic sources (Zhu et al., 2012). The crust–related elements of Mn, Al, Fe, Si, Ca, and Ti mainly result from resuspension and are found bound to PM_{10}. In contrast, elements from anthropogenic sources, such as S, Pb, Zn, vanadium (V), arsenic (As), nickel (Ni), and copper (Cu) were found to be associated with $PM_{2.5}$ (Pekey et al., 2010). Ambient concentrations of Ni, S, Zn, Fe, and K were significantly correlated (generally p <0.02) with their corresponding indoor concentrations. A higher I/O ratio (>1) for Ca, Al, Si, and K indicated contributions from their indoor sources, whereas S, Ni, Zn, and Fe were less influenced by indoor sources (average I/O ratio 0.74–0.83) (Long and Sarnat, 2004). This is in accordance with Molnar et al. (2007) who reported elevated indoor levels of Ca, K, and Ti. Infiltration of ambient Si, S, Mn, Fe, and OC into indoor areas possibly explains the correlation of their concentrations with traffic–related elements (Fe, Mn, Zn, and Br) that were detected indoors (Martuzevicius et al., 2008). Concentrations of the classical tracers of traffic–related air pollution (Pb, Zn, and Br) were substantially higher in areas of high traffic than in low traffic areas (Huang et al., 2007; Olson et al., 2008). Indoor, outdoor, and personal levels of these aforementioned tracers (i.e. Zn, Br, and Pb) were all correlated with each other (Molnar et al., 2006). Some of the traffic–related elements, such as Pb, Mn, and Fe, could also be the result of industrial activities (Minguillon et al., 2012), whereas some (Mn, Cu, Pb) could be tracers of wood burning (Molnar et al., 2005). Levels of crust–related (Al, Fe, Ca, K, and Ti) and automobile–related (Cu, EC, OC, and Zn) elements were higher in vehicles and other microenvironments than levels measured with a fixed–site monitor (Brown et al., 2012).

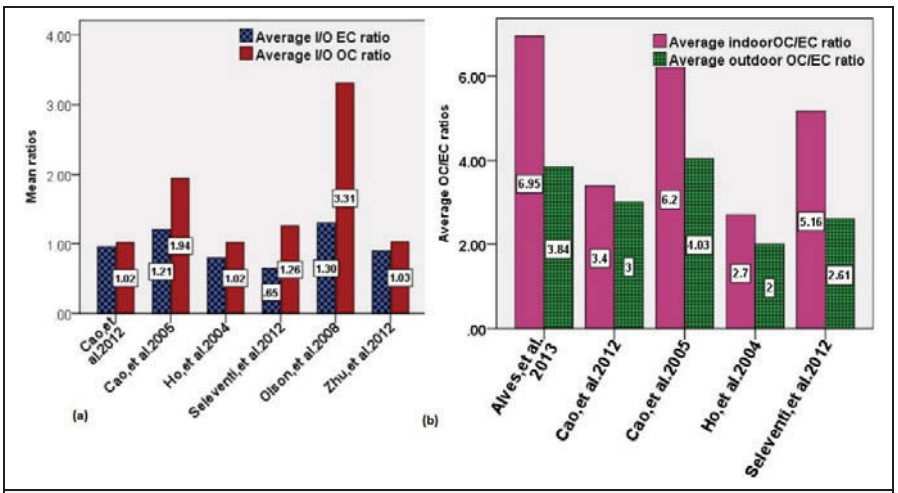

Figure 2. (a) *Average I/O ratios of EC and OC,* ***(b)*** *Average I/O ratios of OC/EC.*

The I/O ratios of Br, Cl, K, Zn, selenium (Se), chromium (Cr), and cobalt (Co) were always >1, primarily because of indoor sources such as environmental tobacco smoke (ETS) and cooking, although there are also ambient sources for these elements (Lim et al., 2011). Adgate et al. (2007) found similar results for Cr, Co, K, and Zn. Prediction of exposure to elements with both indoor and outdoor sources is more complicated than exposure predictions for elements that have only outdoor sources, such as Fe, Mn, and S (Larson et al. 2004). This difficulty in prediction of exposure can be attributed to considerable temporal variations in families' characteristics, personal indoor activities, and other possible factors. Significantly elevated concentrations of S, Ni, Pb, and Br, which are subject to long–range air mass movements, were detected outdoors (Molnar et al., 2007); S, Ni, Si, Mn, Cu, and Fe consistently had higher outdoor concentrations than indoor concentrations (Tovalin–Ahumada et al., 2007).

5. Personal versus Residential Indoor and Outdoor Concentrations

All of the studies evaluated in this section of the review (aside from the study conducted by Crist et al. (2008) in elementary schools) discuss the relationships between residential indoor, residential outdoor, personal, and ambient $PM_{2.5}$ concentrations. Conducting direct exposure assessment through personal measurement rather than indirect assessment can be an appropriate approach for examining exposure to different air pollutants in different settings, reflecting spatial and temporal variations (Cortez–Lugo et al., 2008). However, because of practical reasons such as affordability, time, and willingness of participants to carry measuring tools, researchers have attempted to model personal exposure based on data from fixed–site monitors. Relatively few researchers conducted simultaneous personal, indoor, and outdoor measurements in an attempt to comprehensively assess the total exposure. Higher personal $PM_{2.5}$ mass levels compared to I/O have been demonstrated in several studies (Figures 3 and 4; Adgate et al., 2003; Meng et al., 2005; Adgate et al., 2007; Johannesson et al., 2007; Crist et al., 2008; Minguillon et al., 2012). Higher outdoor levels compared to indoor and personal levels have also been reported (Wheeler et al., 2011).

Conducting multiple simultaneous measurements (i.e., indoor, outdoor, and personal exposure) rather than single measurements in one exposure setting can provide more comprehensive data for describing exposure patterns. For example, Johannesson et al. (2007) found a strong correlation (r=0.90, p<0.0001) between residential outdoor and urban background levels of $PM_{2.5}$. Results of the same study showed also, a strong correlation (r=0.71) was found between personal non–smokers and indoor $PM_{2.5}$, along with a moderate correlation (r=0.56, p<0.003) between personal non–smokers and residential I/O. In such a case, both ambient fixed–site monitors and residential outdoor measurements could give a good estimate of personal exposure, provided that there are no significant indoor sources such as smoking or heating. Minguillon et al. (2012) found similar correlations. Significantly positive correlations were also found between indoor–outdoor and personal concentrations as well as between indoor and outdoor concentrations, but not between ambient background and residential site concentrations (Sorensen et al., 2005). In this case, ambient background monitors (i.e., fixed–site monitors) were not regarded as good estimators of residential outdoor levels (Sorensen et al., 2005). Larson et al. (2004) stated a different view that it may not be easy to predict personal exposure based on only one type of measurement (indoor or outdoor) without conducting source apportionment, in agreement with Meng et al. (2005, 2009) who evaluated contributions of different predictors of personal $PM_{2.5}$ such as personal activities, indoor sources, and indoor–outdoor air exchange rates. For example, Adgate et al. (2003) did not find a strong correlation between personal and ambient background concentrations.

The contribution of ambient PM_{10} to total personal exposure to $PM_{2.5}$ is recorded by Jedrychowski et al. (2006), although study participants spent most of their time indoors and indoor measurements alone may not be enough for estimating exposure to $PM_{2.5}$. Conversely, in another study by Johannesson et al. (2007), PM_1, is found to make up 70–80% of the $PM_{2.5}$ mass concentrations in both ambient and indoor environments. In summary, it is evident that it is more useful to measure particulates within size classes ($PM_{2.5}$, PM_1, and PM_{10}) rather than obtaining measurements of total particulate matter.

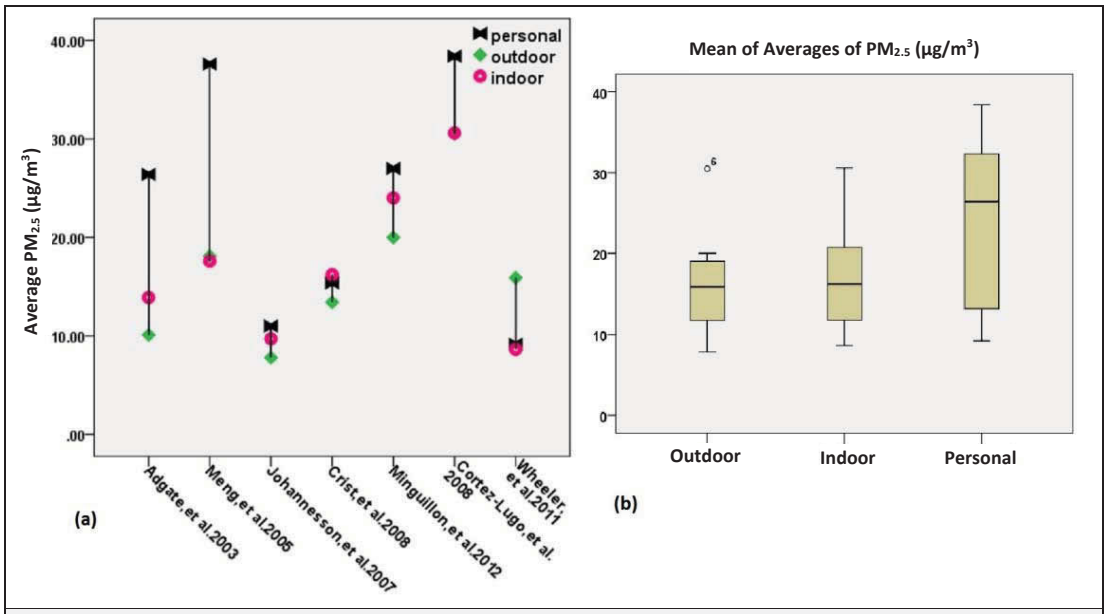

Figure 3. (a) *Personal followed by indoor, then outdoor concentrations of $PM_{2.5}$,* ***(b)*** *General trend of $PM_{2.5}$ mass levels - calculations based on average values.*

Table 1. *Measurements conducted at schools with/without other measuring sites (N=12)*

Study	Measurement Techniques	Country	Sampling Settings	Remarks, Chemical Analysis, & Mass Conc. Analysis
Alves et al. (2013)	Filter–based & real–time	Portugal	Schools	OC, EC, water–soluble ions
Branis et al. (2005)	Filter–based	Czech Republic	Schools	
Castro et al. (2010)	Filter–based	Portugal	Sites close to hospital, schools and universities PAHs, tobacco smoking emissions	
Crist et al. (2008)	Filter–based and real–time	USA	Personal and schools	
Gadkari (2010)	Filter–based	India	Personal and schools	
Fromme et al. (2008)	Filter–based	Germany	Schools, EC, OC, water–soluble ions	
Hassanvand et al. (2014)	Filter–based and real–time	Iran	School dormitory and retirement house	Water–soluble ions
Ho et al. (2004)	Filter–based	China	Office, schools, houses	EC, OC
Huang et al. (2012)	Filter–based	China	Commercial and student dormitory	Water–soluble ions
Molnar et al. (2007)	Filter–based	Sweden	Residential houses and schools	AER, metal analysis
Sawant et al. (2004)	Filter–based	USA	Residential houses and schools	EC, OC, certain organic pollutants
Wichmann et al. (2010)	Filter–based	Sweden	Residential houses and schools	Soot, F_{INF}, and AER

AER=Air Exchange Rate, F_{INF}=Infiltration factor

6. Measurements at Schools

Several studies have investigated children's exposure to $PM_{2.5}$ at schools (Table 1 summarizes those based on active sampling). It is well known that younger children are much more sensitive to air pollution than healthy adults. A large percentage of recent studies on I/O $PM_{2.5}$ have attempted to provide data for this through conducting $PM_{2.5}$ measurements in schools. Active children are likely to be exposed to pollution in different settings, not only in schools. However, conducting $PM_{2.5}$ measurements in schools can provide helpful information that could be useful in estimating the total exposure, particularly if there are enough data on residential indoor–outdoor $PM_{2.5}$.

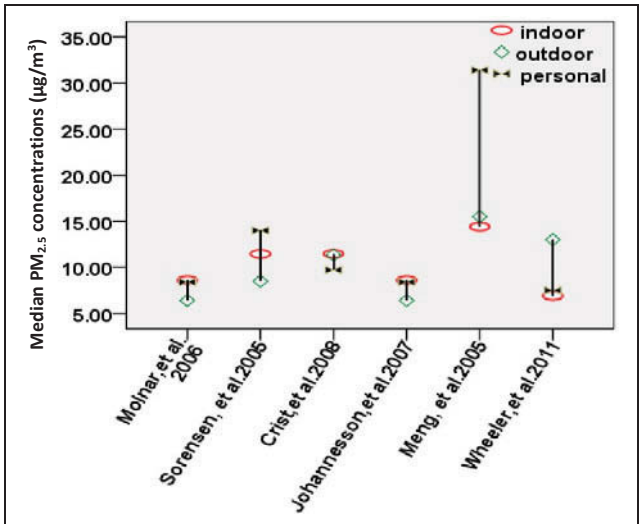

Figure 4. *Personal, indoor and outdoor PM concentrations based on median values.*

Chemical composition and gravimetric analysis of $PM_{2.5}$ indicated that ambient air contributes to high indoor levels of $PM_{2.5}$ in schools and in preschools (Molnar et al., 2007; Sofian and Ismail, 2012; Alves et al., 2013). Hassanvand et al. (2014) estimated that 76% of $PM_{2.5}$ concentrations measured inside of a school dormitory in Tehran (Iran) were of outdoor origin, in agreement with previously reported effects of outdoor $PM_{2.5}$ on indoor $PM_{2.5}$ (Ho et al., 2004). The relatively high proportions of potassium and magnesium reflect the contributions of crust materials on $PM_{2.5}$ in primary schools (Fromme et al., 2008). Significantly high outdoor concentrations of selected elements (S, Ni, Br, Pb) that are attributed to long–range transported air masses have been recorded (Molnar et al., 2007).

It has generally been assumed that increased classroom occupancy will increase the indoor levels of $PM_{2.5}$ through resuspension of aerosols or other possible mechanisms such as students' activities, and use of chalk for writing on the blackboard (Gadkari, 2010; Goyal and Khare, 2011; Alves et al., 2013; Mohammadyan and Shabankhani, 2013), but this is not always the case. For example, Branis et al. (2005), Guo et al. (2010), and Tippayawong et al. (2009) found no significant correlation between $PM_{2.5}$ levels and presence of students indoors. However, positive associations between indoor and outdoor relative humidity and indoor PM levels (including $PM_{2.5}$), and a strong negative correlation between wind velocity and indoor PM (including $PM_{2.5}$); have been found (Branis et al., 2005). Higher levels of $PM_{2.5}$ recorded on the weekends as compared to weekdays, and higher levels recorded during the nighttime than those during the daytime may indicate that indoor activities have less impact on indoor levels of $PM_{2.5}$ (Tippayawong et al., 2009). However, these findings disagree with results of several other studies that showed impact of indoor activities on $PM_{2.5}$ levels (Massey et al., 2009; Cao et al., 2012; Massey et al., 2012). In rare cases, indoor levels of $PM_{2.5}$ are similar to outdoor levels with no observable relationship between ambient air quality and indoor air quality (Wichmann et al., 2010).

7. Sampling Approaches

Short–duration measurements are commonly applied for studying within–day and/or day–to–day variations of $PM_{2.5}$ mass levels. Although the Tapered Element Oscillating Microbalances (TEOMs) and Grimm are among the most widely used samplers for real–time measurements (Mohammadyan and Shabankhani, 2013;

Hassanvand et al., 2014), researchers have recently begun to use a wide variety of samplers that limit the comparability of the results. Among these samplers are DustTrak samplers (Cao et al., 2005), PC–2 Quartz Crystal Microbalance (Kuo and Shen, 2010), the scanning mobility particle sizer, the aerodynamic particle sizer (Long and Sarnat, 2004), and other kinds of particle counters (Tippayawong et al., 2009; Guo et al., 2010; Sofian and Ismail, 2012). Water–based condensation particle counters (Polidori et al., 2007) are useful for the purposes of investigating the daily and within–day variations in $PM_{2.5}$ levels, although such devices are considerably different in their measurement accuracies and sensitivities. Photo–charging (PC) monitors and diffusion–charging (DC) monitors have been suggested for $PM_{2.5}$ measurements (Ott and Siegmann, 2006) while light–scattering based automatic monitors can be used to record temporal variations of indoor $PM_{2.5}$ levels (Alves et al., 2013). Significant day–to–day variability could be observed in personal exposure rather than in outdoor–indoor exposure (Johannesson et al., 2007), and it is expected that $PM_{2.5}$ concentrations during the night would be less than those during the day (Wheeler et al., 2011). However, Branis et al. (2005) did not find a significant difference between daytime and nighttime $PM_{2.5}$ levels.

There are several reasons for discrepancies in the results obtained by real–time devises. Firstly, calibration of each of these devises depends primarily on the calibration technique provided by the manufacturer/suppler [i.e., the "standard" test dust (Alves et al., 2013)]. In contrast, filter–based samplers are commonly calibrated with "primary calibrators" or with flow meters. Secondly, every direct–reading device has an aerosol concentration range (i.e., measuring range). Consequently, very high or very low particle concentrations outside of this range will not be measured accurately. Thirdly, light scattering device (i.e., Grimm) measurements are influenced by the shape of particle. For example, the direct–reading device will give accurate results only if the collected particles are mostly spherical in shape.

Fourthly, some of these direct–reading devices have no clear cut points for particle size fractionation, rather they give approximate estimations of particle concentrations (Kuo and Shen, 2010). Finally, such devices provide two types of readings, either particle number concentration or particle mass concentration, that are considered to be two different units of measurement and are not easily compared (Guo et al., 2010; Mohammadyan and Shabankhani, 2013). In conclusion, real–time samplers are very useful in measuring $PM_{2.5}$ in microenvironments such as schools, houses, and indoor workplaces. However, to get the best results and increase measurement accuracy, it would be ideal to conduct simultaneous measurements with filter–based samplers (normally operated at 1–5 L/min) and with real–time measurements (Kuo and Shen, 2010; Alves et al., 2013; Mohammadyan and Shabankhani, 2013; Hassanvand et al., 2014).

The sampling period for each single sampling session with active sampling is 24 h (Figure 5). Approximately half (51.11%) of the reviewed articles on active sampling adopted a 24 h sampling time in accordance with the international guidelines on exposure to particulate matter which are based on 24 h exposure. Smaller percentages (13.33% and 11.11%) of the reviewed articles adopted 48 h and <8 h, respectively. Some studies adopted a combination of these sampling durations, especially for measurements conducted at schools. In addition, one study adopted multiple sampling durations that exceeded 48 h.

The sampling flow rate must be considered to ensure collection of a large enough volume of air for accurate chemical analysis, and to avoid disturbing the study participants with high noise levels. In addition, shorter periods of sampling would not reflect the total exposure because variations in daily activities and rapid diurnal changes in weather conditions influence ambient $PM_{2.5}$ concentrations. Simultaneous indoor/outdoor and personal sampling flow rates commonly range from 1 L/min – 5 L/min (low flow rate) to avoid noise nuisance generated by the air pumps (Cyrys et al., 2004; Reff et al., 2005; Sorensen et al., 2005; Molnar et al., 2006; Huang et al., 2007; Johannesson et al., 2007; Castro et al., 2010; Kliucininkas et al., 2011; Lim et al., 2011; Wheeler et al., 2011; Cao et al., 2012; Huang et al., 2012; Minguillon et al., 2012; Zhu et al., 2012). However, the U.S. EPA recommended ambient $PM_{2.5}$ sampling flow rate of 16.7 L/min was the second most common flow rate used in the reviewed studies (Ramachandran et al., 2003; Molnar et al., 2005; Molnar et al., 2006; Johannesson et al., 2007; Pekey et al., 2010; Kliucininkas et al., 2011; Lim et al., 2011). Other studies used a flow rate of 10 L/min (Ramachandran et al., 2003; Cyrys et al., 2004; Long and Sarnat, 2004; Hovorka et al., 2005; Reff et al., 2005; Molnar et al., 2007; Olson et al., 2008; Wichmann et al., 2010). A very small percentage of studies used flow rates not included within the above ranges. The use of Harvard impactors could indicate or lead to the reappraisal of the accuracy and popularity of these samplers for $PM_{2.5}$ measurements (Hovorka et al., 2005; Molnar et al., 2005; Molnar et al., 2007; Martuzevicius et al., 2008; Olson et al., 2008; Wichmann et al., 2010).

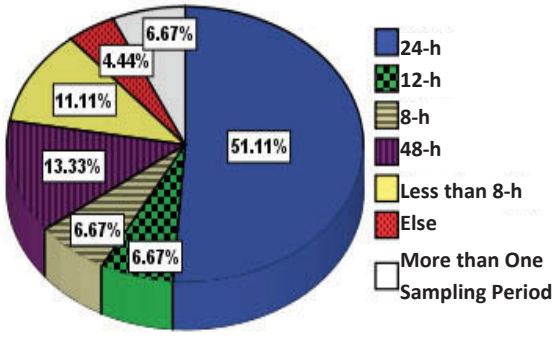

Figure 5. *Sampling durations for single sampling sessions adopted in the (45) studies on active sampling.*

8. $PM_{2.5}$ Modeling

Several studies have intensively examined the infiltration factor (F_{INF}) in an attempt to evaluate the effect of ambient air on the indoor environment (Ho et al., 2004; Long and Sarnat, 2004; Molnar et al., 2007; Polidori et al., 2007; Fromme et al., 2008; Gadkari, 2010; Guo et al., 2010; Lim et al., 2011; Hassanvand et al., 2014). For all reviewed studies, the average value of F_{INF} was greater than 0.40 (except in Wichmann et al. 2010 where F_{INF}=0.25) and in most cases it exceeded 0.60, indicating high infiltration of outdoor $PM_{2.5}$ into indoor environments (Figure 6; Hanninen et al., 2004; Molnar et al., 2007; Fromme et al., 2008; Martuzevicius et al., 2008; Sangiorgi et al., 2013). The determination of the I/O ratio of sulfate/sulfur concentrations has been an acceptable approach for estimating F_{INF}, provided that there are no significant indoor sulfur sources (Hanninen et al., 2004; Long and Sarnat, 2004; Fromme et al., 2008; Allen et al., 2012; Hassanvand et al., 2014; Hanninen et al., 2004; Sawant et al., 2004; Reff et al., 2005; Huang et al., 2007; Martuzevicius et al., 2008; Olson et al., 2008).

The strong correlation between indoor and outdoor levels of sulfur indicates the insignificance of indoor sulfur sources and supports the use of sulfur as a tracer for calculations of infiltration of ambient air into indoor areas (Hassanvand et al. (2014) (r=0.87–0.94); Fromme et al. (2008) (r=0.70); and Long et al. (2004) (r=0.91)). Irrespective of the amount that ambient air infiltrates into indoor areas, the variations in F_{INF} values between sampling sites may reflect variations in exposure to concentrations of $PM_{2.5}$ originating outdoors (Allen et al., 2012; Minguillon et al., 2012). These variations in F_{INF} have been attributed to differences in air

exchange rates (Hanninen et al., 2004; Long and Sarnat, 2004; Meng et al., 2005), ambient temperature (Meng et al., 2009), and ventilation systems (Wichmann et al., 2010).

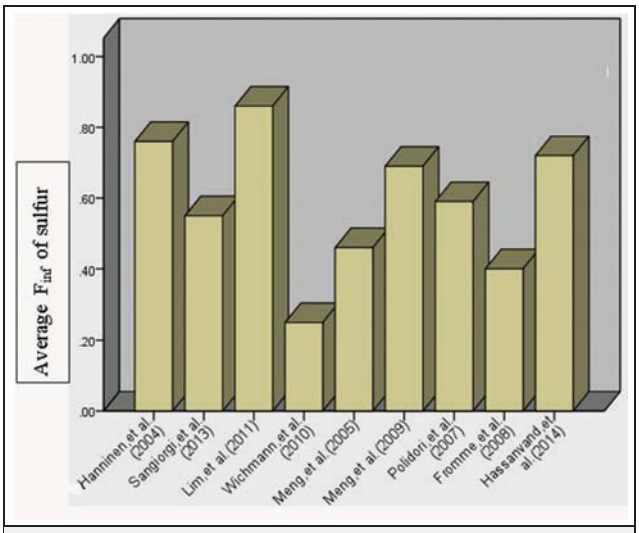

Figure 6. *Average values of infiltration factors (F_{inf}) of sulfur.*

The majority of reviewed studies that investigated the contribution of outdoor air into indoor areas used sulfur/sulfate to calculate F_{INF}. However, a small number of studies have estimated F_{INF} using other tracers such as Nickel (Long and Sarnat, 2004), EO, and OC (Polidori et al., 2007). Allen et al. (2012) successfully examined a new approach for predicting F_{INF} that could be used in large–scale studies of exposure assessment. Both the conventional approach for estimating F_{INF} using indoor/outdoor ratios of sulfur and the recursive model (RM) that was previously developed and validated were used by Polidori et al. (2007) in their study on I/O carbonaceous contents of $PM_{2.5}$ measured in retirement homes.

The trend or trajectory of personal exposure to $PM_{2.5}$ varies considerably between individuals (Larson et al., 2004; Gerharz et al., 2009) and could be estimated by portable GPS devices and a record of daily personal activities. However, this type of modeling needs to be supported by data from actual outdoor measurements (Gerharz et al., 2009).

Residential outdoor $PM_{2.5}$ concentrations could be estimated by using information gathered by background ambient monitors (Baxter et al. 2007; Johannesson et al., 2007). Residential outdoor $PM_{2.5}$ concentrations are commonly correlated with residential indoor $PM_{2.5}$ concentrations. Therefore, total personal exposure could be estimated without conducting personal measurements, which may occasionally be infeasible or inapplicable. In contrast, Adgate et al. (2007) found that the ambient monitors underestimate both personal and residential indoor levels of trace elements. However, as personal exposures vary considerably, a type of source apportionment would be helpful to identify the contribution of individual pollution sources to personal exposure.

Receptor models such as Positive Matrix Factorization (PMF) and Chemical Mass Balance (CMB) were adopted in a limited number of the reviewed studies. PMF is a useful approach for PM source apportionment (Larson et al., 2004; Minguillon et al., 2012). Using PMF, Minguillon et al. (2012) identified six indoor/outdoor sources of $PM_{2.5}$ (sulfate, sea salt, fuel oil, road traffic, industrial, cigarette, and mineral) whereas Larson et al. (2004) identified five different sources (crustal, vegetation burning, sources rich in chlorine, mobile sources of emissions, and secondary sulfate). Although these two studies were somewhat similar in their methods, the results were slightly different, possibly because of spatial variations in the outdoor sources.

There has been recent substantial interest in using the mass balance–based models for source apportionment. Goyal et al. (2011) developed and statistically confirmed the usefulness of a single compartment mass balance–based model for predicting indoor concentrations of particulates (including PM_{10}, $PM_{2.5}$,and PM_1) in naturally ventilated classrooms, under the assumption of isothermal conditions. Meng et al. (2005) used another single compartment mass balance–based model to estimate the contribution of outdoor sources to indoor concentrations of $PM_{2.5}$ at 56%. When determining the mass of indoor $PM_{2.5}$ of outdoor origin, variables such as human activities (e.g., use of air conditioning or open windows) should be considered (Hodas et al., 2014).

Enrichment factor analysis, defined by Huang et al. (2007) as "the average element concentrations of the indoor and outdoor particle relative to the elements of the Earth's crust", has long been used for source apportionment. It has been used for differentiating the natural sources of PM from several anthropogenic sources. Based on enrichment values, the measured chemical elements are classified into enriched, moderately enriched, and non–enriched. The elements S, Se, As, Br, Pb, CL, and Zn are classified as highly enriched and Cr, Cu, Ni, V are classified as enriched, indicating that they have anthropogenic sources which can be specifically identified.

9. Miscellaneous Studies

Although several studies have estimated the potential harmful health effects of exposure to $PM_{2.5}$, a limited number of the reviewed studies estimated the expected health benefits and improvements in air quality from applying different air conditioning systems or indoor configurations. Multi–zone indoor air quality modeling has shown that the proper use of efficient in–duct air filters can reduce premature deaths, hospital visits and asthma). The application of whole–house air cleaners can have positive public health effects (MacIntosh et al., 2010). Cortez–Lugo et al. (2008) conducted a study of indoor, outdoor, and personal exposure to $PM_{2.5}$ by individuals suffering from COPD (chronic obstructive pulmonary disease) who were expected to spend most of their time indoors. The results showed significant contributions of indoor sources to total personal exposure to $PM_{2.5}$, implying the necessity of conducting measurements indoors rather than outdoors. In a study on the exposure of pregnant women to particulate matter, ambient PM_{10} and ETS have contributed significantly to total personal exposure to $PM_{2.5}$ (Jedrychowski et al. 2006). Indoor–outdoor relationships between both $PM_{2.5}$ and PM_{10} are frequently observed (Cortez–Lugo et al., 2008; Massey et al., 2009; Kuo and Shen, 2010; Pekey et al., 2010; Goyal and Khare, 2011; Massey et al., 2012; Hassanvand et al., 2014). Temporal variations have been detected in $PM_{2.5}$ levels but not in PM_{10} levels (Kliucininkas et al., 2011). Indoor concentrations of both $PM_{2.5}$ and PM_{10} were significantly affected by outdoor concentrations of $PM_{2.5}$ and PM_{10} (Fromme et al., 2008; Kuo and Shen, 2010; Pekey et al., 2010; Hassanvand et al., 2014). High levels of indoor human activity and space heating are also possible contributors to high indoor concentrations of $PM_{2.5}$ and PM_{10} in the winter (Massey et al., 2012).

10. Limitations

- This review focused on the general trends in research interests and all articles that fulfilled the inclusion criteria are discussed, regardless of the sample sizes, *p*–values or analysis methods.
- This review focused only on studies of indoor–outdoor $PM_{2.5}$, and studies on health effects were intentionally excluded.
- Studies on particulate matter in general (rather than on only $PM_{2.5}$) were excluded, although such studies may contain information on indoor–outdoor $PM_{2.5}$.
- Studies on active sampling and studies on real–time monitoring were reviewed, but not equally. This review concentrated

primarily on filter–based studies because of the comparable results.

11. Conclusions

This literature review consistently found correlations between indoor and outdoor $PM_{2.5}$ and it is evident ambient air contributes to indoor $PM_{2.5}$ levels. Several studies examined and determined the relevance of using sulfate/sulfur to calculate F_{INF}. High average values of F_{INF} indicated high infiltration of outdoor $PM_{2.5}$ into indoor environments, although variations in F_{INF} between sampling sites were reported within the same study. Sources of $PM_{2.5}$ including smoking, infiltration of vehicle exhaust, coal and oil combustion, road dust, heating in winter, and building materials contributed to the higher indoor $PM_{2.5}$ levels, compared to outdoor levels, that were reported in more than 40% of the reviewed studies. Effective education programs are needed to control outdoor $PM_{2.5}$ sources (i.e., traffic and industry) and to increase the awareness of the general public about use of efficient energy, public transport, and other efforts that may reduce emissions of $PM_{2.5}$, and therefore indoor $PM_{2.5}$ concentrations. Regulatory agencies must also apply green policies and effective regulations. In this regard, a paramount question emerges: how could governments' rules be optimized to make actual decisions? Further studies need to be conducted to propose effective control measures for outdoor sources of $PM_{2.5}$.

The majority of studies on active sampling used 24 h sampling periods at flow rates ranging from 1–5 L/min and also at 16.7 L/min, as recommended by the U.S. EPA. Harvard impactors were popular samplers for active sampling.

A large percentage (42%) of the reviewed filter–based studies were conducted in Europe, followed by 38% in the USA, and 20% in Asia, indicating a gap in $PM_{2.5}$ research in other parts of the world. Among the reviewed filter–based studies, no research was conducted in Africa, Latin America, Russia, or Australia. Therefore, certain aspects associated with $PM_{2.5}$ measurements, such as the chemical composition, the air exchange rates (that in turn influence the infiltration into indoor areas), and other issues reported in these studies, could not be globally generalized. However, the findings of the reviewed studies remain essential for determining general trends in PM studies. Based on the reviewed articles, it is difficult to conclude that ambient fixed–site monitoring could give accurate estimations of actual total exposure because of contradictory results.

The search terms used to obtain the articles from the ScienceDirect and Web of Science databases removed many studies on health effects from exposure to $PM_{2.5}$. The majority of the articles in this review focused on indoor–outdoor $PM_{2.5}$ levels and exposure in residential houses and school settings; other exposure settings such as in hospitals and industries were not considered. The expected health benefits and improvement in air quality from applying different air conditioning systems or indoor configurations and ventilation modes was reported only in four of the reviewed articles. A very limited number (≤2%) of studies in this review investigated possible air quality improvements resulting from different air conditioning systems or indoor configurations. In addition, pollution levels in slums, where people use different types of fuels for cooking, have not been given adequate consideration.

The abundance of OC in indoor $PM_{2.5}$, which was reported in some papers, was attributed to outdoor sources. Indoor concentrations of PAHs were attributed to outdoor sources such as vehicle exhaust in addition to the indoor sources of tobacco and wood smoke. Water–soluble ions were suggested to originate from outdoor sources. Sulfate, nitrate, and ammonium were the greatest contributors to both indoor and outdoor $PM_{2.5}$ mass concentrations, indicating their abundant sources. Studies that measured Ca, Al, and K had contradictory results as to whether these crust–related elements have significant outdoor or indoor sources. However, Ca was found to greatly contribute to indoor $PM_{2.5}$ mass concentrations.

12. Recommendations

- In the near future, it will be important to conduct studies on exposure to $PM_{2.5}$ in rural areas because of the many complex sources of exposure within modern cities. Rural areas can be used as control sites for urban areas and can provide useful information for source apportionment.
- Analysis of sulfate and/or sulfur concentrations is important in indoor/outdoor studies of $PM_{2.5}$ because they are consistently related to outdoor concentrations, which supports their relevance in infiltration factor calculations.
- Several studies have examined whether or not ambient monitors provide accurate estimations of residential outdoor concentrations or personal exposure to $PM_{2.5}$. However, it is still unclear under what conditions these monitors provide optimal estimation; this needs further investigation.
- The use of sulfate concentrations for F_{INF} calculations, along with the analysis of Ca and K concentrations, will help identify contributions of both indoor and outdoor $PM_{2.5}$ sources.
- Because of strong evidence of the infiltration of outdoor $PM_{2.5}$ into indoor environments, further studies are needed to propose effective control measures for outdoor sources.
- Moreover, further studies are needed to address the topic of health benefits and improvements in air quality beside topics such as the pollution levels in residential houses in suburbs and slums where poor people depend mainly on natural ventilation. To generalize the research findings, additional studies need to be conducted in Russia, Africa, and some parts of Asia.

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Supporting Material Available

Summary of studies based on active sampling with/without other techniques (Table S1). This information is available free of charge via the internet at http://www.atmospolres.com.

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