

Review Article

Indoor aerosols: from personal exposure to risk assessment

Abstract Motivated by growing considerations of the scale, severity, and risks associated with human exposure to indoor particulate matter, this work reviewed existing literature to: (i) identify state-of-the-art experimental techniques used for personal exposure assessment; (ii) compare exposure levels reported for domestic/school settings in different countries (excluding exposure to environmental tobacco smoke and particulate matter from biomass cooking in developing countries); (iii) assess the contribution of outdoor background vs indoor sources to personal exposure; and (iv) examine scientific understanding of the risks posed by personal exposure to indoor aerosols. Limited studies assessing integrated daily residential exposure to just one particle size fraction, ultrafine particles, show that the contribution of indoor sources ranged from 19% to 76%. This indicates a strong dependence on resident activities, source events and site specificity, and highlights the importance of indoor sources for total personal exposure. Further, it was assessed that 10–30% of the total burden of disease from particulate matter exposure was due to indoor-generated particles, signifying that indoor environments are likely to be a dominant environmental factor affecting human health. However, due to challenges associated with conducting epidemiological assessments, the role of indoor-generated particles has not been fully acknowledged, and improved exposure/risk assessment methods are still needed, together with a serious focus on exposure control.

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Practical Implications

The indoor source contribution can be the dominant fraction of the integrated daily residential exposure to particles (especially UFP and PM_{2.5}), strongly depending on resident activities, source events, and site specificity. Up to 30% of the burden of disease from particulate matter, exposure can be attributed to indoor-generated particles, which are thus one of the dominant environmental factors affecting human health globally. Improved exposure and risk assessment methods are needed, together with a serious focus on exposure control, to reduce the severity and risks associated with human exposure to indoor particulate matter.

Introduction

Throughout their entire lives, each and every person is exposed to the aerosols omnipresent in indoor air. Given that most people spend the majority of their lives indoors, the consequences of this exposure range from insignificant to fatal and depend on the type of aerosols present, type of indoor environment, duration of time spent, age, gender, susceptibility, and many other factors (Adgate et al., 2003; Brasche and Bischof, 2005; Lai et al., 2004; Leech et al., 2002). With the increasing contemporary focus on indoor exposure, it is worth noting that it has been known for centuries that exposure to certain types of aerosols constitute at least some level of risk and should be controlled (Miller, 1998).

Considering the scale of the problem and the potential severity of the associated risks, the need for good characterization and quantification of exposure to indoor aerosols appears obvious. Yet, while the past decade has seen an increase in the body of literature published on this topic, there are still major challenges to be addressed, to fully understand and quantify the magnitude of both individual and population exposure to air pollution in different types of indoor microenvironments. There are several reasons for this. Firstly, *exposure* is often confused with *concentration*, and it is pollutant concentration, not human exposure that is usually characterized or quantified. It does not help that the term ‘*exposure*’ belongs equally in the domain of everyday language and in the scientific vocabulary, and in the former it does not have a precise meaning. It is this vague understanding of *exposure* that is often adopted in indoor sciences. Secondly, ambient particulate matter (PM) presents a special challenge, and it has not even been established conclusively which of its characteristics are the most significant in relation to health. Thirdly, apportioning indoor exposure to the two components essential for exposure control, including: (i) PM of outdoor origin (which has penetrated indoors) and (ii) PM generated by indoor sources, is a complexity with which many studies have struggled. These three aspects are discussed in more detail below.

Exposure is a vital element of risk assessment, a process which is initiated upon identification of a hazard,

and evidence that exposure constitutes risk to human health. While a detailed discussion of this process is outside the scope of this review, and more information on the Predictive Risk Equation can be found in USEPA (1991) or Naugle and Pierson (1991), it is very important to understand the terms used and the individual elements of this process. In particular, *pollutant concentration* is the numerical value of the amount of an individual pollutant per unit volume of air at a particular point in time or averaged over a period of time. *Exposure* is a product of the *pollutant concentration* and the *time* over which a *person* is in contact with that pollutant. When concentration varies with time, the time-averaged concentration is used for exposure calculation. Two types of exposures are of special importance in health risk assessments: (i) lifetime exposure, which is the sum of exposures which occurred in different environments – this is particularly important for carcinogenic pollutants; and (ii) short-term exposure to elevated concentrations. *Dose* is a product of *exposure* and *dosimetry factors* (such as inhalation rate, regional surface area of the lung or breathing pattern), and quantifies the amount of substance available for interference with metabolic processes or biologically significant receptors. *Dose-response* is the magnitude of the response of an individual to a given dose of a pollutant. *Lifetime individual risk* (which applies specifically to carcinogenic pollutants) is a product of *lifetime average daily dose* and *dose-response*. *Risk to exposed population* is a quantitative assessment which takes into account *lifetime individual risk* of the population groups affected and specific *exposure scenarios* of the population. The main objective of health risk assessment is to link the hazard with the risk to the exposed population in a quantitative way, and thus provide the basis for risk management.

In the context of this discussion, exposure assessment includes: (i) identification of sources of indoor pollution and specifically PM; (ii) characterization (through measurements, surveys, questionnaires, etc) of indoor air pollutants and contributing factors such as concentration, duration, frequency, distribution route, time frame, and geography; and (iii) modeling exposure for different occupancy and lifestyle scenarios. To quantify personal exposure to PM, three methods are frequently used: (i) direct personal exposure

measurements, which are the best and most accurate measure of exposure (Jantunen et al., 2002); (ii) measuring microenvironmental concentrations and the time spent in these microenvironments; and (iii) personal activity information (Meng et al., 2005). The latter involves gathering personal activity information from questionnaires and has been used in both regression and analysis of variance models to describe sources and activities impacting exposure and indoor air quality (Baxter et al., 2007; Koistinen et al., 2001).

Regarding the second of the above listed methods, the existing exposure models adopt one of two general approaches: (i) a time series approach that estimates the microenvironmental exposure of individuals based on the instantaneous concentration at any moment; or (ii) a time-averaged approach that estimates the microenvironmental exposure using average pollutant concentrations and the total time spent in each microenvironment (USEPA, 2004). Expressions for the two exposure evaluation methods are described in Equations (1) and (2), respectively:

$$E(t_1, t_2) = \int_{t=t_1}^{t=t_2} C_s(t) dt \quad (1)$$

$$E(t_1, t_2) = \bar{C}_s(t_2 - t_1) \quad (2)$$

where t_1 and t_2 denote the start and stop times of the investigated time interval; $E(t_1, t_2)$ is the exposure during this period; $C_s(t)$ is the instantaneous microenvironmental concentration at any moment between t_1 and t_2 ; and \bar{C}_s is the average pollutant concentration during the time period $t_2 - t_1$. The units of both equations are [*pollutant concentration* \times *time*].

It is noteworthy that different definitions have been used for exposure, and for example, Zartarian et al. (1997) defined exposure as the contact between an agent and a target that takes place at a contact boundary over an exposure period. According to their definition, time-averaged inhalation exposure is in units of *concentration* (at the breathing zone boundary), while integrated exposure is in units of *concentration* \times *time*. To avoid confusion, if there is only concentration data available, we propose that one should refer to the *concentration to which a person is/would be exposed*. However, in the case of actual exposure data, which always has a duration (with the minimum duration being one single breath), then the units should be: [*pollutant concentration* \times *duration of exposure*]. Otherwise, it would be difficult to avoid confusion between ‘time-averaged inhalation exposure’ and ‘integrated exposure’ (over time), with each having different units.

Exposure to particles is a special challenge, due to their multifactorial nature and their highly variable size. Airborne particles range in size over five orders of

magnitude, from molecular dimensions to the sizes that are distinguishable with the naked eye (from about 0.001 μm to about 100 μm ; Baron and Willeke, 2001), which constitutes a big challenge in the choice and availability of instrumental techniques, particularly for personal exposure monitoring. Other characteristics include: number concentration, number size distribution, mass concentration mass size distribution, surface area, shape, chemical composition, electrical charge, or light-scattering properties. Usually, only some of these properties are measured, and almost never all of them, simultaneously.

In relation to some of these metrics, in particular number concentration of ultrafine particles (UFP $< 0.1 \mu\text{m}$) or their surface area, while there is ample evidence of the hazardous nature of these particles provided by toxicological studies, epidemiologic studies have not yet established an exposure–response relationship (Fissan et al., 2007; Shin et al., 2007; WHO, 2005). In contrast, for $\text{PM}_{2.5}$ and PM_{10} (mass concentration of particles with aerodynamic diameters smaller than 2.5 and 10 μm , respectively), and total suspended particulate matter (TSP), not only has their hazardous nature been established, but epidemiologic studies have quantified exposure–response relationships (WHO, 2005). However, whether the effects are independent or dependent and therefore only some of them need to be measured (and regulated) are open questions at present. Thus, in the absence of a well-established exposure–response relationship, the selection of parameters for investigation is not a trivial aspect.

Indoor and outdoor source contribution to indoor exposure to aerosols

Indoor particles are a mix of ambient particles that have infiltrated indoors, particles emitted indoors and particles formed indoors through reactions of gas-phase precursors emitted both indoors and outdoors (Meng et al., 2005; Morawska and Salthammer, 2003; Uhde and Salthammer, 2007). Therefore, the composition and toxicity of indoor particles is very complex, with similarities but also differences to outdoor aerosols. These differences reflect particle origin, as well as postformation physicochemical processes, in both indoor and outdoor environments. Epidemiological associations between PM and various health outcomes are based predominantly on ambient air measurements (WHO, 2005); however, poor correlations have been found between ambient PM concentrations and personal exposure to PM. This is due to the contribution of many other pollution sources to personal exposure, including work related, in-vehicle and/or leisure activities (Meng et al., 2005). This initiated a debate as to whether ambient PM is a good surrogate for exposure to PM and in some recent work assessing personal exposure to PM, a division of personal exposure into

ambient and nonambient sources has been considered (Wilson and Brauer, 2006; Wallace and Williams, 2005).

Infiltration of outdoor particles is significantly modified by particle size-specific differences in the penetration efficiency and indoor deposition rate. The latter also directly affects the concentrations generated by indoor sources. Both coarse and ultrafine particles are rapidly removed from the air, but by different mechanisms: coarse particles by gravimetric settling, and ultrafine (e.g., those generated by gas cooking or candles) by thermokinetic deposition. Accumulation mode particles, which dominate outdoor background concentrations, have the highest penetration and infiltration rates (Long et al., 2000). To account for particles which infiltrated from outside, indoor to outdoor ratios for particle concentration are often calculated (I/O). Many studies have reported I/O ratios both with and without operating indoor sources, and on the basis of comparisons between several published articles, Morawska and Salthammer (2003) concluded that, for naturally ventilated buildings in the absence of indoor sources, I/O ratios for PM₁₀ and PM_{2.5} ranged from 0.5 to 0.98 (with a median value of 0.7) and 0.54 to 1.08 (median, 0.91), respectively. This highlights the importance of the contribution of outdoor air as a source of particles encountered in indoor environments. However, when indoor sources are present, I/O ratios for PM₁₀ and PM_{2.5} range from 1.14 to 3.91 (median, 1.47), and 1 to 2.4 (median, 1.21), respectively, demonstrating the significance of indoor source contributions.

Indoor sources of ultrafine particles [contributing to particle number (PN), rather than mass concentration] have been identified as tobacco smoking, cooking (frying, grilling, baking, barbequing, boiling water, toasting, cooking soup, etc), the use of gas and electric stoves, electric toasters, gas-powered clothes dryers, candle and incense burning, photocopiers, laser printers, hair spray, cleaning products, and furniture polish containing terpenes (which form secondary aerosols when in the presence of ozone) (Abt et al., 2000; Dennekamp et al., 2001; He et al., 2004; Hussein et al., 2006; Lee and Hsu, 2007; Li et al., 1993; Long et al., 2000; Ogulei et al., 2006; Wallace, 2005, 2006; Weschler, 2003; Wainman et al., 2000). Larger indoor particles (contributing to particle mass concentration, PM_{2.5} and/or PM₁₀) have been identified as coming from sweeping, hovering, dusting, human movement (walking, dancing, children playing), sitting on upholstered furniture, resuspension from clothes (folding clothes, blankets, making a bed), washing powder residues (zeolite), resuspension from carpets and the pouring of kitty litter (Abt et al., 2000; Ferro et al., 2004; Gudmundsson et al., 2007; Koistinen et al., 2004; Long et al., 2000; Ogulei et al., 2006).

Some of these sources, such as frying (Abt et al., 2000;) or physically stirring foods that are sautéing in fats and liquids (Long et al., 2000), can contribute to both the smaller and larger particle concentrations. It has been reported that indoor sources can periodically generate particles that, in terms of number and mass concentrations, significantly exceed background levels (Abt et al., 2000; He et al., 2004; Long et al., 2000) and that indoor particle events are site and time specific, and can also be brief, intermittent and highly variable (Hussein et al., 2005; Long et al., 2000; Nazaroff, 2004). A phenomenon which also needs to be mentioned in relation to human exposure is the 'personal cloud'. As discussed by Wallace (1996), the personal cloud effect is an observed elevation in personal exposure when measured by a personal monitor relative to indoor concentration measured by a fixed room monitor. While the exact nature of this phenomenon has not been explained, a review by Wallace (1996) has revealed that it can elevate personal exposure by up to 50%.

Aim and scope of this work

The aim of this study was to review the existing literature on personal exposure to indoor aerosols and the risk it poses to the following: (i) identify the state of the art in experimental techniques used for personal exposure assessment; (ii) compare the exposure levels reported by studies conducted in different settings and in different countries; (iii) assess the overall role of outdoor background versus indoor sources in contributing to personal exposure; and (iv) examine scientific understanding of risk due to personal exposure to indoor aerosols. The special focus of the review was on exposure in domestic and school environments due to their importance to overall human exposure. Exposure in other important microenvironments, such as in-transit, has already attracted separate reviews (Knibbs et al., 2011), or are sufficiently complex and diverse to warrant separate reviews (in particular industrial and non-industrial workplace, healthcare facilities, or facilities manufacturing or handling engineered particles). In addition, not included in this review were studies reporting indoor concentrations or exposures in developing countries, where wood, coal, agricultural residues, dung, and biogas are used as fuels, because differences in household characteristics, type of fuel, cooking appliances and ventilation conditions make comparisons difficult. Another exclusion were environments affected by environment tobacco smoke. In developing countries, indoor exposure to particles from burning biomass fuels is a major health concern, and the reported concentrations are substantially higher in comparison with those reported in developed countries, with indoor PM₁₀ shown to range from 104 to 2500 $\mu\text{g}/\text{m}^3$ and PM_{2.5} 201–304 $\mu\text{g}/\text{m}^3$ in developing

countries (Begum et al., 2009; Morawska et al., 2011a; Wang et al., 2010). Again, this is a topic for a separate review.

The authors of this review identified studies published in English, between January 1989 and October 2012, using ScienceDirect, EBSCOhost, Web of Science and Wiley Interscience search engines. The following key words were used: personal exposure, indoor environments, PM₁₀, PM_{2.5}, PM₁, and UFP. Additional studies were identified from the references of these publications, and on the basis of personal knowledge of the authors of this review.

Methods for monitoring personal exposure to indoor particulate matter

General approaches to PM monitoring for the purpose of personal exposure assessment include the following: (a) measurements carried out in outdoor microenvironments, usually at fixed outdoor stations, to assess compliance with national ambient air quality standards. As discussed above, this approach fails to account for all components of exposure and is also a poor predictor of personal exposure to outdoor PM, due to its large spatial and temporal variations, especially in urban environments (e.g., Kousa et al., 2002); (b) monitoring carried out simultaneously at indoor and outdoor sites (Williams et al., 2003), including single or multiple indoor microenvironments (Rodes, 2011). In such studies, the sampler inlet is usually placed 1.0–1.5 m above the floor to simulate a seated breathing height, and in a location not unduly influenced by potential nearby aerosol sources (Adgate et al., 2002); and (c) direct personal exposure monitoring.

Table S1 summarizes the existing methods that have been used for PM characterization for approaches (a) and (b), as discussed above, including particle mass (Almeida et al., 2011; Andresen et al., 2005; Braniš et al., 2005; Crist et al., 2008; Ekmekcioglu and Keskin, 2007; Jo and Lee, 2006; Landis et al., 2001; Lee et al., 2002; Monn et al., 1997; Morawska et al., 2001, 2003; Reff et al., 2007; Williams et al., 2000), PN (Bhangar et al., 2011; Diapouli et al., 2007; He et al., 2007; and Hoek et al., 2008; Marra et al., 2010; Mattson, 2004; Morawska et al., 2001, 2003; Mullen et al., 2011a; Wallace and Ott, 2011), surface area (Buonanno et al., 2010) and size distribution measurements (Almeida et al., 2011; Buonanno et al., 2010; Liao et al., 2006; Marple, 2004; Massey et al., 2012; Morawska et al., 2001; Saraga et al., 2011; Wan et al., 2011; Williams et al., 2003), as well as that of particle composition (Chao and Wong, 2002; Hoek et al., 2008; Janssen et al., 2001, 2003; John et al., 2007; Lai et al., 2004; Landis et al., 2001; Molnár et al., 2007a,b; Reff et al., 2007; Roorda-Knappe et al., 1998; Stranger et al., 2008; Titcombe and Simcik, 2011; Williams et al., 2003; Wilson et al., 1995). It should be noted

that this is not an exhaustive review of all available methods – for a comprehensive review of particle monitoring methods the reader is directed to Morawska et al. (2008). In general terms, the methods can be divided into online and off-line techniques. Some of these methods are suitable for both indoor and outdoor application, while others are more appropriate for outdoor environments (due to high flow rates or the excessive noise they generate). It can be seen from Table S1 that there is a large range of instrumentation available for particle mass concentration measurements, which is not surprising because many types of instruments are necessary for standard compliance monitoring. Inertial impactors have been used since 1860 to collect particle samples for chemical and gravimetric analysis (Marple, 2004). The cascade impactor was developed in the 1940s, and today, there are precision cascade impactors for high accuracy aerosol sampling to collect size-fractionated airborne particle samples. For example, the 125B Rotating Micro-Orifice Uniform Deposit Impactor (MOUDI) from MSP Corp is designed for high concentration aerosol sampling in 13 stages, from 10 nm to 18 μm (MSP Corp, 2011). Once collected on an impactor or filter, the particulate mass is determined by thermally equilibrating and weighing the sample. Subsequently, chemical analysis may be carried out by several techniques such as atomic absorption spectrometry, atomic fluorescence spectrometry, inductively couple plasma spectroscopy, and X-ray fluorescence spectroscopy (XRFS). Particle mass concentration is not only monitored by the classic method of collecting particles on filters, but it is increasingly monitored using online methods, in particular photometers, which calibrate aerosol light-scattering responses for a specific challenge aerosol to provide real-time mass concentration readings (Rodes, 2011). Light-scattering techniques have been applied to monitor size-fractionated mass concentrations such as PM₁₀, PM₅, PM_{2.5}, and PM₁ (Massey et al., 2012). It can be further concluded from Table S1 that in addition to several instruments available for PN concentration and size distribution monitoring, there are two PN personal monitors and only one instrument for surface area monitoring. In consequence, there is very little information available to assess personal exposure to surface area, with only a handful of investigations conducted in occupational settings (Wang et al., 2010), including one in a pizzeria (Buonanno et al., 2010). Today, there are a number of techniques available for collecting samples for elemental and carbon analysis [organic carbon (OC) and elemental carbon (EC)] and a selection of these are listed in Table S1.

While microenvironment monitoring can provide estimates of personal exposure, accurate results require the use of monitoring devices that can be carried on or by the participants, for direct personal exposure monitoring [point (c) above]. This is important to avoid mis-

classification, noncompliance, and to improve the fidelity of the data in general. The first large-scale probability-based study of personal exposure to particles was conducted in 1990 by the Harvard University School of Public Health (HSPH) who designed and constructed a battery-operated personal monitor for both PM_{10} and $PM_{2.5}$ (Ozkaynak et al., 1996). The monitor was worn over 24 h by 178 participants. The two major indoor particle sources were found to be smoking and cooking. However, even in the presence of these sources, the majority of indoor particles came from outdoors. Yet, indoor concentrations were only weakly correlated with outdoor concentrations, and the correlation between personal exposures outdoor concentrations was even weaker.

Hand-held ultrafine particle monitors are now commercially available, including the TSI 3007 Condensation Particle Counter (CPC) (Wallace and Ott, 2011). Recent advances in the miniaturization of optical benches used for nephelometry have produced compact real-time aerosol sensors that can be worn or carried to characterize acute exposure levels (Rodes, 2011). Two real-time PN and size monitoring devices that are being used at present are the Nanotracer Monitor from Philips (Marra et al., 2010) and the Mini Diffusion Size Classifier (DiSC) from the University of Applied Sciences, Windisch, Switzerland (Fierz et al., 2007). In June 2012, Philips ceased production of Nanotracers, however, the DiSC is still commercially available. In addition, Naneos Particle Solutions, Switzerland, is presently introducing a pocket size/postcard size particle detector, providing the lung-deposited surface area of aerosol particles with a time resolution of 5 s (Naneos, 2012). These personal exposure monitors are small, self-contained, battery-powered devices that can be carried by an individual to mimic the proximity of the breathing zone, defined as 30 cm in diameter around the head, to local sources or spatial concentration gradients (Jensen and O'Brien, 1993). Contemporary personal aerosol exposure monitoring may be direct reading or time integrated sampling systems. They generally consist of two parts – (i) the aerosol sizing and collection unit, which includes the sample inlet, an impactor or a cyclone for aerodynamic sizing and the filter cassette holder, if applicable, and (ii) the flow control, electronics and sensors unit, which includes a mini-pump and battery pack for the purpose of flow management (power management, flow control, timing functions, and start/stop functions). Some devices may be fitted with built-in supplementary features for compliance and activity level sensing, Global Positioning System sensing, QC data capture and sensors for temperature and humidity. Typically, participants carry the personal samplers in small foam-insulated bags with a shoulder strap that has the inlet mounted on the front (Broich et al., 2011). During sampling sessions, participants are requested to wear or carry the expo-

sure monitors whenever possible and to place them beside them while seated or sleeping (Adgate et al., 2002; Jayaratne et al., 2011). During sampling, participants are also asked to fill out a time–activity diary, recording the time periods spent in primary microenvironments such as at home, outside the home, inside other than home, outside other than home, and in-transit. They also record data on exposure to tobacco smoke and other potential modifiers of exposure, such as occupation, cooking, outdoor activities, and the number of hours that doors and windows were open in a residence (Mazaheri et al., 2012). There have been many investigations of personal exposure conducted with portable devices and, in Table 1, we present several such examples.

Personal exposure levels

Residential environment

People in developed countries spend approximately 65% of our time in our residences (Brasche and Biscohof, 2005; Leech et al., 2002). Studies that assess concentrations measured in residences, outdoors, concentrations from personal monitoring and integrated daily residential exposure are given in Table S2. Residences with smoking have been excluded, as tobacco smoking is a known source of fine particles that can dramatically influence indoor concentrations. For example, indoor $PM_{2.5}$ mass concentration was reported to increase by 58–130% due to smoking (Breysse et al., 2005; Stranger et al., 2007). Studies assessing the particle concentration and emission factors of a given indoor activity or cooking event have not been included in this review. In the reviewed studies (Table S2), simultaneous indoor and outdoor concentration data are most commonly given. Fourteen studies reported integrated mass concentration values from direct personal monitoring with varying averaging times (note that in the original articles these values are presented as ‘personal exposure concentrations’ but given that exposure is a product of concentration and exposure duration, and for consistency within this review, we will refer to them as ‘concentrations from personal monitoring’) for $PM_{2.5}$ (Adgate et al., 2002; Johannesson et al., 2007; Koistinen et al., 2004; Lai et al., 2004; Landis et al., 2001; Meng et al., 2005; Molnár et al., 2005; Rodes et al., 2010; Turpin et al., 2007; Wallace et al., 2006), $PM_{1.5}$ (Williams et al., 2000) and PM_{10} (Lioy et al., 1990; Monn et al., 1997). Only three studies give values for daily residential exposure to UFP in units of number concentration per hour per day (Bhangar et al., 2011; Mullen et al., 2011b; Wallace and Ott, 2011). Average $PM_{2.5}$ from personal monitoring in the listed studies ranges from 10.6 to 54 $\mu\text{g}/\text{m}^3$, with average and median values of 27.3 and 26.5 $\mu\text{g}/\text{m}^3$, respectively. These

Table 1 Summary of personal exposure monitoring studies using portable devices

Metrics	Size	Instruments	References
Particle mass	PM ₁₀	Personal Environmental Monitor (SKC)	Scapellato et al. (2009)
		Grimm Aerosol Spectrometer 1.109	Broich et al. (2011)
	PM _{2.5}	Battery-operated personal monitor (Harvard School of Public Health)	Ozkaynak et al., 1996;
		Personal Environmental Monitor model 200 (MSP Corp)	Andresen et al. (2005)
		Filter Sampler URG-2000-25F (URG)	Crist et al. (2008)
		PM _{2.5} Personal sampler (BGI)	Reff et al. (2007)
		Personal environmental monitoring sampler (PEMS) (Harvard School of Public Health)	Jedrychowski et al. (2006)
		SidePak Portable nephelometer model AM510 (TSI)	Borgini et al. (2011)
	PM ₁	Battery-operated personal monitor (Harvard School of Public Health)	Ozkaynak et al., 1996;
		Grimm Aerosol Spectrometer 1.109	Broich et al. (2011)
Particle number and average size		Personal exposure monitor PMON (Model URG-2000-15, URG)	Williams et al. (2000)
		Nanotracer Monitor (Philips)	Jayarathne et al. (2011)
			Mazaheri et al. (2012)
			Buonanno et al. (2012a)
Particle surface area		MiniDiSC (Windisch)	Fierz et al. (2007)
Particle size distribution		Nanoparticle Dosimeter (Naneos Particle Solutions)	Naneos (2012)
Elemental composition	PM _{2.5}	Not available	
Black carbon	PM _{2.5}	GK2.05 cyclone sampler (KTL, BGI)	Lai et al. (2004)
		Personal Environmental Monitor (PEM) (MSP)	Landis et al. (2001)
Light scattering	TSP	Reflectometer EEL 43	Roosbroeck et al. (2007)
Polycyclic Aromatic Hydrocarbons	PM _{2.5}	Nephelometer MIE pDR	Williams et al. (2003)
		Portable nephelometer SidePak model AM510 (TSI)	Borgini et al. (2011)
		Personal microenvironment Aerosol Speciation Samplers PMASS model 240 (MSP Corp)	Titcombe and Simcik (2011)

concentrations are given for different averaging times, including 24 h, 48 h, or 7 days, incorporate time spent outdoors, and comprise both adults and children. Bhangar et al. (2011), Mullen et al. (2011a,b), and Wallace and Ott (2011) present an elegant way of calculating indoor residential (i.e., while occupants are at home) daily integrated exposure to UFP in units of particles per cm³·h/day. The daily integrated exposure is a normalized form of integrated exposure and may serve as a useful single metric for comparative purposes. The average indoor residential daily integrated exposure per person reported in these three studies ranged from 11.5×10^4 to 29.6×10^4 particles/cm³·h/day. On the basis of values reported by Wallace and Ott (2011), the average integrated daily residential exposure (cooking + indoor background due to infiltration of particles from outdoors) constitutes 67% of the total daily integrated personal exposure. Additionally, in these three studies, each occupant's daily integrated residential UFP exposure was apportioned into contributions from outdoor, episodic and continuous indoor sources, on the basis of continuous indoor and outdoor measurements, occupant activity logs and questionnaires. The indoor source contribution to the average residential daily UFP exposure was 59% (ranging from 38% to 76% in seven residences) in Bhangar et al. (2011) and 30.5% (ranging from 19% to 42% in two residences) in Mullen et al. (2011a,b). Recalculating values given by Wallace and Ott (2011), cooking (dominant indoor source) contributed to 47% of residential exposure (one residence). Wallace (2006) attributed 55% of particles found indoors to indoor

sources. These studies point to a strong dependence on resident activities, source events and site specificity, but also highlights the importance of indoor sources in total personal exposure.

In Figure 1, the statistics (minimum values, 1st quartile, median, 3rd quartile, and maximum values) obtained from average values reported in the reviewed studies are given for outdoor and indoor PM₁₀ and PM_{2.5}, and for PM_{2.5} from personal monitoring. The purpose of Figure 1 is to give an indication of the range of measured concentrations, given that the applied averaging times varied to a great degree, as did the measurement methods and instrumentation used. For example, the averaging times ranged from hours (8, 24, 48 h) to days, seasons and years, and some included nonoccupancy time. In terms of assessing personal exposure to particles in residences, the most relevant average would be during the time the occupant is present in the residence (occupancy time). Inclusion of nonoccupancy time (when no-one is present in the residence) underestimates the concentration relevant for personal exposure assessment. Median values of reported indoor PM₁₀ concentrations are slightly higher than outdoors (i.e., 34.7 and 30.2 µg/m³, respectively), while median indoor and outdoor values for PM_{2.5} are the same (17.6 µg/m³), with higher variations seen indoors. The median value of PM_{2.5} from personal monitoring accounts for 26.5 µg/m³ and is higher than indoor and outdoor concentrations. This can be explained by the fact that people spend time in other microenvironments where concentrations can be higher than at indoor and outdoor measurement sites

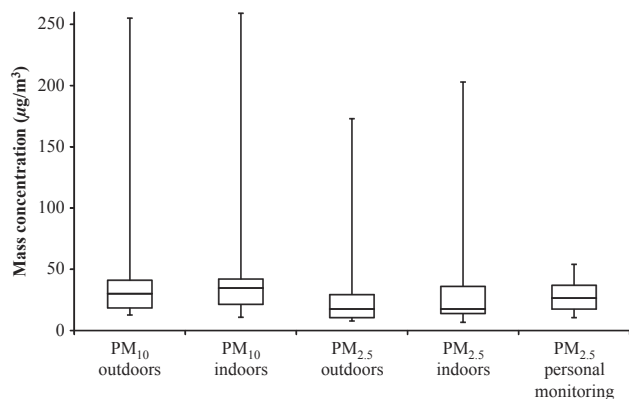


Fig. 1 Summary of particle mass (PM_{10} and $PM_{2.5}$) obtained from average values reported in the studies included in Table S2. Box plots denote minimum values, 1st quartile, median, 3rd quartile and maximum values

(e.g., smoky bar, restaurant, in-transit, woodworking site, etc) or they can be in closer proximity to indoor sources than the indoor monitor (Meng et al., 2005; Turpin et al., 2007).

Eight studies assessing indoor and outdoor PN concentrations were found and have been included in Table S2. However, due to differences in minimum measured size (between 6 and 15 nm) and the variety of averaging times (based on the whole monitoring period, occupancy, activity, and nonactivity periods), their direct comparison did not seem meaningful. Only two studies (Bhangar et al., 2011; Mullen et al., 2011a,b) reported directly measured values for occupancy time (while awake and asleep) and used instruments with the same lower size limit (6 nm). On the basis of these two studies (seven single family houses and four apartments), the average concentration for occupancy time indoors accounts for 16.1×10^3 (ranging from 5.3×10^3 to 34.7×10^3) particles/cm³, while outdoors accounts for 19.0×10^3 (ranging from 8.9×10^3 to 22.4×10^3) particles/cm³.

Similarity of the indoor to outdoor concentrations (in Figure 1 and the text above) may be misleading and may suggest that there is not a pressing need to supplement ambient air-based exposure proxies with a more detailed assessment of personal exposures. However, when comparing indoor and outdoor concentrations, one has to bear in mind that: (a) the buildings filter a substantial fraction of outdoor particles (i.e., infiltration of particles originating from outdoors is significantly modified by particle size-specific differences in the penetration efficiency and indoor deposition rate), thus modifying exposure to ambient particles; (b) in addition to particles of an ambient origin, particles can also originate from indoor sources, and these may differ significantly from outdoor particles in terms of both composition (as they have different sources) and temporal patterns; (c) personal exposure is further modified by both outdoor (e.g., in

traffic) and indoor (e.g., in kitchen) near-field activities, which may not be captured properly by stationary microenvironmental monitoring.

Some activities, including smoking, woodworking, and cooking, were reported to have a dramatic influence on fine indoor PN concentrations (Hussein et al., 2006; Wallace et al., 2006) and on personal $PM_{2.5}$ exposure (Meng et al., 2005; Turpin et al., 2007; Wallace et al., 2006). Reported peak number concentrations due to cooking activities ranged from 1.6×10^4 to 6.3×10^5 particles/cm³ (He et al., 2004; Wan et al., 2011), and these are much higher than reported outdoor maximum concentrations. Sources of indoor-generated particulate matter are not well characterized, with contributions from a range of vastly different activities, including cooking, cleaning, combustion devices, candles, photochemistry, printers, and the use of various consumer products such as aerosols, detergents, sprays, etc.; however, their quantitative contributions have not been determined on a population representative basis.

In addition, the impact of particle size-dependent physical processes has not been sufficiently investigated, in terms of their impact on PM exposure and uptake, particularly when particle composition is considered in addition to physical characteristics. For example, particles of different chemical composition tend to have characteristic size distributions, and it is not known whether the differences observed in composition studies (e.g., Levy et al., 2012) are only partly or completely attributable to the physical characteristics. To determine whether the observed differences may also be attributable to composition or exposure, and whether dose differences are due to particle size distributions, requires new research applying innovative approaches to exposure and dose characterization, based on integrated monitoring and modeling approaches. This is discussed further in section 'Chemical'.

Epidemiological studies attempting to investigate the health effects of exposure to indoor-generated particles are typically limited to small cohorts or very inaccurate questionnaire-based exposure assessment. European data on indoor concentrations of indoor- and outdoor-generated particles showed that while the outdoor component ranged from 6 to 20 $\mu\text{g}/\text{m}^3$, the contribution of indoor sources in non-smoking homes was 3–5 $\mu\text{g}/\text{m}^3$, representing 20–30% of the total concentrations (Hänninen et al., 2004).

Schools

The focus of this section is personal exposure assessment in relation to airborne particle concentrations in schools, in particular PM_{10} , $PM_{2.5}$, and UFP. Schools are complex indoor environments with very specific building designs, ventilation conditions, indoor and outdoor pollution sources, and types of activities

conducted (Daisey et al., 2003; Lee and Chang, 2000). Therefore, the monitoring of school exposures is challenging and can be conducted in many different ways, as reported by the recent reviews on this topic (Lin and Peng, 2010; Mejía et al., 2011). On the other hand, the characterization of these microenvironments is considered important to estimate the total exposure of children. Children are a population subgroup very sensitive to air pollution, because they receive a higher dose of airborne particles relative to lung size compared with adults (Farhat et al., 2005), and at the same time, their physiological and immunological systems are still in the process of developing.

Generally speaking, air quality in schools can be measured on three scales, depending on the spatial unit of analysis (Mejía et al., 2011): (i) 'city scale', the broadest and most common scale characterizes air quality across several city blocks using remote measurements; (ii) 'school scale', which characterizes air quality in schools from data collected by ground-level monitors or samplers installed in school buildings, school yards and/or around the perimeter of the school; or (iii) 'personal scale', which measures air quality by attaching individual portable instruments to children, to assess their exposure.

Within this review, a literature search was conducted for articles reporting data relevant for assessing the air quality in and around schools. The most important criterion for inclusion was that the collected air quality data were used for indicating or representing pollutant concentrations in school environments. In the past decade, there has been a large body of literature published on the concentration levels of airborne PM in school classrooms, specifically PM_{10} and $PM_{2.5}$. Data extracted from about 40 original papers are summarized in Table S3.

A number of studies examining PM levels in classrooms, mainly in Asia and Europe, reported high levels of PM_{10} and $PM_{2.5}$ (Almeida et al., 2011; Borgini et al., 2011; Diapouli et al., 2008; Ekmekcioglu and Keskin, 2007; Fromme et al., 2005, 2007; Goyal and Khare, 2009; Janssen et al., 1997; Lahrz et al., 2003; Lee and Chang, 2000; Liu et al., 2004; Roorda-Knape et al., 1998; Stranger et al., 2007, 2008; Yang et al., 2009), ranging from 30 to $1181.1 \mu\text{g}/\text{m}^3$ and 13 to $360 \mu\text{g}/\text{m}^3$, respectively. Other studies carried out in the United States and Northern Europe (Brunekreef et al., 1997; Gauvin et al., 2002; Janssen et al., 2001, 2003; John et al., 2007; Keeler et al., 2002; Link et al., 2004; Molnár et al., 2007a; Parker et al., 2008; Shaughnessy et al., 2002; Weichenthal et al., 2008) reported lower levels of PM_{10} (average value $23 \mu\text{g}/\text{m}^3$) and $PM_{2.5}$ ($14 \mu\text{g}/\text{m}^3$) than other published literature (Table S3). The wide range of PM_{10} and $PM_{2.5}$ concentrations indicate a great potential for their reduction, as well as a need for identifying the factors responsible for this variability.

On the other hand, there are only a few studies reporting on indoor UFP concentrations in school classrooms, and even less which investigated the correlation with outdoor traffic or indoor processes (Buonanno et al., 2013; Diapouli et al., 2008; Guo et al., 2008; Morawska et al., 2009a; Mullen et al., 2011a,b; Weichenthal et al., 2008). Only one paper reported the daily personal exposure of children and the contribution of the school microenvironment to daily dose (Buonanno et al., 2012a). In general, classroom UFP concentrations decreased with a reduced degree of traffic density and urbanization, indicating that, in the absence of significant indoor sources, vehicular emission greatly influenced the indoor UFP concentration levels.

It should be noted that articles included in this review varied in their design and approach, because personal exposure assessment was not the main focus of all studies. The main differences can be summarized as follows: (i) the averaging time (school time, 24 h, 3 weeks, etc); (ii) selection of schools (random or based on defined conditions); (iii) the spatial unit of analysis (city, school or personal scale); (iv) identification of the major sources of local air pollution and the corresponding thermodynamic processes (some techniques include meteorological or local traffic data); (v) statistics (summary, correlation, principal component, and time series analyses are commonly applied in the schools); and (vi) analysis of the main parameters influencing indoor concentrations (building age, seasons, ventilation rates, and indoor activities).

In Figure 2, the statistics (minimum values, 1st quartile, median, 3rd quartile and maximum values) of the particle mass (PM_{10} and $PM_{2.5}$) and number concentrations obtained from the studies included in Table S3 are reported. In terms of PM_{10} , the median value for all schools was significantly higher indoors ($102 \mu\text{g}/\text{m}^3$) than outside the schools ($37 \mu\text{g}/\text{m}^3$). This increment is largely due to indoor sources like resus-

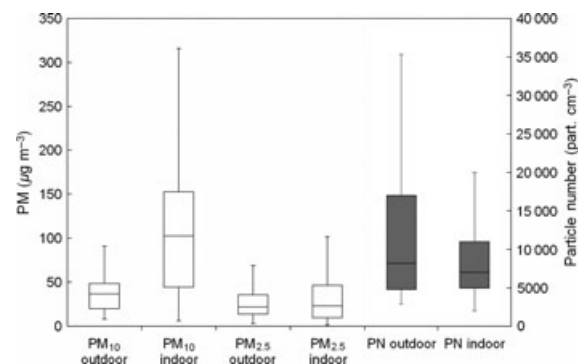


Fig. 2 Distribution of the particle mass (PM_{10} and $PM_{2.5}$) and number concentrations obtained from the studies included in Table S3

pension, which mainly influence the coarse fraction (Braniš et al., 2005; Kingham et al., 2008). The corresponding wide range of indoor PM₁₀ (higher than outdoor PM₁₀) indicates an important potential for reduction and the need to determine the main influential parameters for this variability. This confirms that, to assess the personal exposure of children in schools, indoor PM₁₀ measurements are vital, along with the complete characterization of indoor sources and children's activities.

PM_{2.5} data were similar for both indoor and outdoor sampling sites, with a median value equal to about 23 µg/m³. This variability is slightly higher in indoor environments, confirming a reduced influence of indoor activities (mainly resuspension) on the fine fraction.

In Figure 2, negligible differences between the outdoor and indoor measurements (8.2×10^3 particles/cm³ and 7.0×10^3 particles/cm³, respectively) are also reported for P) concentrations. This is due to the major influence of UFP sources (mainly traffic) on outdoor monitoring sites, where, in the absence of relevant indoor UFP sources, the building envelope smothers the 'fresh' outdoor peaks. Therefore, to assess the personal exposure of children in schools to UFPs, outdoor measurements are very important and have to be included in the experimental design. In terms of personal exposure, the data were limited to particle mass concentration measurements reported in three of the reviewed papers (Borgini et al., 2011; Gauvin et al., 2002; Janssen et al., 1997), and therefore, it was insufficient for performing any statistical analysis. It should be noted that the data presented in Figure 2 do not account for differences in how the data were averaged, with the concentrations reported in Table S3 being based on a variety of averaging times (school time, daily, weekly, etc). Therefore, the main conclusion that can be drawn from the data presented in Figure 2, that indoor PM₁₀ and UFP concentrations are higher and lower than outdoor levels, respectively, would be further enhanced if only data for school times were considered.

To assess personal exposure, data are needed that characterize concentrations when a specific person is present in a given space. Concentrations given as daily or weekly averages are not suitable, as these concentrations capture times when pupils are not at school, and consequently, they are generally lower than specific school time concentrations.

From data available in the reviewed articles, it can be concluded that, in classrooms, PM₁₀ concentrations during school hours were higher than outdoor concentrations, whereas UFPs significantly increased with increasing truck traffic density and significantly decreased with increasing distance from the road. In general, the scientific literature is largely insufficient with respect to: (i) monitoring personal exposure to

particles in schools; ii) UFP monitoring (limited to few papers); iii) concentrations based on an averaging time that only captures when the person in question is present in a given space (i.e., children are at school); iv) the relationship between ambient concentration and personal exposure; and v) the contribution of school exposure to a child's daily exposure, with respect to other sources (transportation, home, outdoors, etc).

Particle composition

Chemical

As discussed in the previous chapters, particulate matter in the indoor environment may be generated by many different indoor and outdoor sources. The composition of airborne particles is generally subject to considerable time and location-related fluctuations, due to these differing types of emission sources, as well as different atmospheric conditions (Turner and Colbeck, 2008). Particles are generally composed of a few main, and many trace constituents, and the predominant chemical components are sulfate, nitrate, ammonium, sea salt, minerals, OC, and EC (Pöschl, 2005).

Air exchange between a buildings indoor and outdoor environment can lead to changes in the mixture of particles found in indoor air, and this process often leads to correlations between indoor and outdoor concentrations. For instance, Geller et al. (2002) found indoor/outdoor (I/O) correlations in EC, sulfate, aluminium, silicon, calcium, iron, titanium, zinc, and potassium for desert aerosols in California. However, these correlations are not always strong, and the composition of indoor and outdoor air particles can differ greatly, as demonstrated by Conner et al. (2001), in a study conducted in Baltimore, MD in the United States. In addition to building air exchange rate, the following processes influence the I/O ratio for the different chemical species in particles:

- Particle penetration from outdoors through mechanical system ventilation ducts, or through cracks in windows, doors, and building walls (Nazaroff, 2004).
- Particle generation due to primary sources, such as from combustion processes, heating, cooking (Evans et al., 2008), household activities (Géhin et al., 2008), hobbies, mechanical wear, biogenic substances, skin, hair, pets, etc.
- Particle generation or alteration due to secondary impacts, such as partitioning of semivolatile organic compounds (SVOCs) and chemical processes such as secondary organic aerosol (SOA) formation.
- Particle deposition and resuspension.

In many outdoor particles, elements and metals can be found which allow an insight into their origin. For instance, iron, silicon, aluminium, calcium, and potas-

sium usually originate from natural sources, and tin, lead and transition metals are generally anthropogenic. Also, certain compounds tend to exist predominately in particles of particular sizes. Anlauf et al. (2006) analyzed inorganic aerosol components as a function of the particle aerodynamic diameter. Ammonium and sulfate tended to be present in groups with a smaller diameter, and nitrate, sodium, and chloride were found more often in the larger particle groups. The distribution of metals in aerosols was investigated by Allen et al. (2001). Cadmium, tin, lead, and selenium tended to be found in smaller particles, while nickel, zinc, copper, cobalt, manganese, and mercury were spread across a wide range of sizes, and iron, strontium and barium tended to be found more in larger particles. Karanasiou et al. (2007) examined aerosols in the city air in Athens for nine metals in different particle sizes. Anthropogenic components, such as cadmium and vanadium, were found in smaller size groups, while copper, chrome, iron, and aluminium were generally found in the larger aerosols. There were no clear tendencies for lead, nickel, and manganese, which can also be attributed to anthropogenic sources. Extensive investigations into heavy metal pollution in airborne particles have been carried out by Amato et al. (2009) and Klumpp et al. (2009).

Given that these outdoor particles infiltrate into the indoor environment, these relationships often hold for indoor particles as well. For instance, Chao and Wong (2002) demonstrated the dependency of elemental composition on particle size for $PM_{2.5}$ and PM_{10} in indoor air. Indoor processes affect this composition as well, with both Morawska et al. (2009a,b) and Barthel et al. (2011) finding small quantities of metals in aerosols that were emitted by laser printers. Grgic (2008) also summarized papers concerning the analysis of metals in aerosols in a review article, which stated that the concentration of metals in aerosols and their bioavailability (Voutsas and Samara, 2002) is influenced by a plethora of factors, such as the pH value, aerosol type and size, organic material, and EC content (Desboeufs et al., 2005).

The particles produced by indoor sources can have completely different compositions depending on their origin. For example, while OC and EC are always released in combustion processes, Hedberg et al. (2002) found that polycyclic aromatic hydrocarbons (PAH), aluminium, silicon, phosphor, sulfate, and chlorine are also released when burning beech wood in a household oven. Burning candles were examined by Pagels et al. (2009) and depending on the product, phosphate, sulfate, sodium, and potassium were found to be significant aerosol components, besides EC and OC. Another strong source of emissions is incense sticks, which are often burned for religious reasons. Depending on the type of incense burned, the particles can contain large quantities of EC, OC, anions, and

main, and transition elements (See and Balasubramanian, 2011).

Nanosprays are a relatively new particle source in indoor environments. Norgaard et al. (2009) analyzed the aerosols from a number of different sprays used for cleaning surfaces and found high levels of cyclic and perfluorinated siloxanes in the air both during and after their use. Many sprays contain silver as an antibacterial active ingredient, and Quadros and Marr (2011) found that the emission of silver correlated with that of chlorine in many of the spray aerosols. The particles released by photocopiers do not, as previously speculated, consist of toner carbon (Wensing et al., 2011), but they mainly arise through condensation of SVOCs. Similarly, Wensing et al. (2008) found silicon and higher alkanes (C31–C45) in printer-generated aerosols, which indicates secondary formation processes, and it is also assumed that the particles found to be released by household electrical appliances arise via SVOCs released by heat (Schripp et al., 2011).

In addition to this, chemical reactions between unsaturated organic substances and ozone take place indoors in a similar way to which they do in atmospheric chemistry and lead to the formation of SOAs. Monoterpenes, such as d-limonene and α -pinene, are usually involved in this process, and these compounds originate from indoor and outdoor sources, due to the use of consumer products and wood off-gassing, respectively. Ozone/monoterpene reactions result in many compounds, including: reactive intermediates such as hydroxyl radicals, alkylperoxy radicals, and Criegee biradicals; high volatility products such as carbon dioxide or formaldehyde; and semivolatile products such as carboxylic acids and hydroperoxides, which may yield SOAs (Kroll and Seinfeld, 2008). Indoors, SOA formation may be due to nucleation or gas-to-particle partitioning, and indoor formation influences particle distributions in the ultrafine and fine size ranges. Enhanced particle formation also may occur via polymerization reactions of the oxidation products within the particle phase. Particular analysis was performed on the SOA formation potential of natural paints (Lamorena et al., 2007), wood products and natural paints (Schripp et al., 2012; Toftum et al., 2008), air fresheners (Lamorena and Lee, 2008), household products (Coleman et al., 2008), and controlled terpene mixtures (Waring et al., 2011).

Few studies have ascertained the personal exposure of building occupants to different compositional elements of indoor particles, and those that have, analyzed integrated gravimetric filter samples. For instance, Zhao et al. (2006b) collected 24-h personal exposure samples for 38 individuals in four different environments and attributed various sources to levels of exposure to different compounds. The composition of indoor particles was a function of the building air exchange rate, with indoor sources dominating the

fractional contribution to the composition at lower air exchange conditions. Sulfur, iron, lead, and EC were mainly attributed to outdoor sources (e.g., vehicle emissions), so the presence of these compounds increased with the air exchange rate. The most extensive study to measure personal exposure to particle composition was the RIOPA study, which measured outdoor, indoor, and personal exposure to particles and volatile organic gases for 100 residents within 100 different homes in three US cities (Weisel et al., 2005). Functional group and elemental analysis were performed on the personal exposure samples. The RIOPA study inferred that personal exposures were frequently modified by indoor sources that generated organic material with a strong aliphatic character (Turpin et al., 2007).

Biological

Bioaerosols contain an heterogeneous mixture of particles from microorganisms (fungi, bacteria, viruses), plants and animals (Dillon et al., 1996). The size of these particles varies significantly, ranging from viruses (20–300 nm) to typical bacterial cells (0.5–3 μm), fungal spores (1.5–30 μm), and pollen grains (10–400 μm) (Reponen et al., 2001). In the literature, bioaerosols most often refer to fungal and bacterial aerosols, but in indoor air, it also includes particles from house dust-mites, cockroaches, and insects and pets, as well as skin cells from humans and pets (Flannigan, 2001).

In general, outdoor air is the main source of fungal bioaerosols, and therefore, the tightness of a building's envelope and the performance of filtration in a ventilation system will determine the I/O ratio in a given building, which is expected to be less than one. However, in areas where the ground is frozen or covered with snow at various times throughout the year, indoor sources become the most significant contributor to indoor bioaerosol concentrations. In these situations, a high I/O does not necessarily indicate the presence of exceptional sources and exposure. Many human activities can affect bioaerosol concentrations, such as handling firewood, root vegetables and plants, which have been shown to elevate fungal spore concentrations (Hunter et al., 1988; Lehtonen et al., 1993). In relation to bacteria, humans themselves can be an important indoor source (Nevalainen 1989; Qian et al., 2012), and humans or pets can also carry bioaerosols from the outdoors (Lehtonen et al., 1993) or from highly contaminated environments, like cow barns (Pasanen et al., 1989). Resuspension also elevates indoor bioaerosol concentrations, during activities such as cleaning indoor surfaces.

In most studies, personal exposure to bioaerosols is assessed indirectly by measuring the concentration of bioaerosols in occupied rooms or spaces. This indirect

method has been found to underestimate exposure to both fungal and bacterial bioaerosols (Wang et al., 2012). One possible explanation for this the role of human occupancy as a source of indoor biological aerosols. For example, Qian et al. (2012) reported size-resolved, per person-hour emission rates for biological particles based on a mass-balance modeling approach and illustrated the extent to which being in an occupied room results in exposure to bacteria that are associated with previous or current human occupants. Recently, the qPCR methodology was developed to assess viruses in the air, but like other methods, it does not give precise information about personal exposure (Ziros et al., 2011). Fragments or constituents measured in dust samples have also been used as an indicator for assessing exposure to dust-mite allergens (Pauli et al., 1988).

One group of researchers used Button Inhalable Samplers to measure the personal exposure of teachers to bioaerosols during winter in Finland. The overall average particle mass concentration was found to be 57 $\mu\text{g}/\text{m}^3$, with a total fungi count of 12.2×10^3 spores/ m^3 and 33 viable fungi colony-forming units/ m^3 . Corresponding values in the home and workplace (classroom) were 17 $\mu\text{g}/\text{m}^3$, 10.8×10^3 spores/ m^3 and 30 cfu/ m^3 , and 34 $\mu\text{g}/\text{m}^3$, 12.0×10^3 spores/ m^3 and 19 cfu/ m^3 , respectively. Bacteria concentrations in the classroom (14.5×10^4 spores/ m^3 , 1090 cfu/ m^3) were found to be higher than in the home (60.6×10^3 spores/ m^3 , 340 cfu/ m^3), as well as higher than overall average personal exposure (86.0×10^3 spores/ m^3 , 720 cfu/ m^3) (Toivola, 2004; Toivola et al., 2002, 2004). Fungi concentrations were generally higher in the classrooms with a higher number of pupils, as well as in homes with dogs that lived inside or those that reported the occasional condensation of water of inner window surfaces. Personal exposure to bacteria was found to be higher for men than women and for people in younger age groups (27–40 and 40–48) compared with older ones (>48 years) (Toivola, 2004; Toivola et al., 2004).

From the point of view of health outcomes, exposure assessment to causative agents is also important. Some of these are present in microbial cell walls (β -glucans) and can be analyzed as whole spores and also as fragments of cell walls. Adhikari et al. (2013) assessed the endotoxin and fungal fragments in 15 homes using cyclone samplers which divided particles into three ranges: <1.0 μm , 1.0–1.8 μm , and >1.8 μm . According to enzyme activity and limulus amoebocyte lysate tests, the <1.0 μm particles contributed up to 63% (mean, 22.7%) and 96% (mean, 22.6%) of activities in enzyme activity and endotoxin responses, respectively. Miniature cyclone sampling methods to assess personal exposure to bioaerosols have also been developed. However, cyclones samplers often cause stress for bacteria and may compromise their viability when

compared to stationary liquid impingers (Tolchinsky et al., 2011).

Intake/uptake and deposition in the lungs

Lung deposition modeling

In the indirect dose assessment approach, the daily PN or surface area deposited dose in tracheobronchial and alveolar airways for a given age group can be computed as: the product of the inhalation rate of a given age group depending on the human activity; the fractional deposition depending on inhalation rate and particle diameter, integrated over the whole PN size distribution; and the time spent for a given activity in a defined location (Buonanno et al., 2011, 2012b). Thus, to relate the inhaled ambient aerosol to bronchial and alveolar doses requires knowledge of the fractional deposition efficiencies of inhaled particles in different regions of the human respiratory tract. Particle deposition is determined by biological factors, such as lung morphology and breathing patterns, and physical factors such as fluid dynamics, particle properties and deposition mechanisms (Hofmann, 2011).

At present, the direct experimental *in situ* determination of particle deposition in human subjects is limited to total deposition during a single breath for a wide range of particle sizes and flow rates (Hofmann et al., 2009; Löndahl et al., 2006, 2008, 2009; Morawska et al., 2005; ICRP, 1994; Heyder et al., 1986). Bronchial and alveolar deposition as a function of particle size and flow rate can only be derived indirectly and with less accuracy from subsequent retention measurements of radiolabeled aerosols (Heyder et al., 1986) or from the analysis of serial bolus deposition data (Kim and Hu, 1998). However, health risk assessment for inhaled particles requires information on local deposition patterns within the lungs and such information can only be provided by computational modeling.

Current particle deposition models can be grouped into two categories, referring to the region of interest in the lung: (i) deposition in the whole lung (whole-lung models); or (ii) deposition in a localized region of the lung (local-scale models) (Hofmann, 2011). In whole-lung models, particle deposition in individual airways is computed by analytical equations for particle deposition efficiencies under specific flow conditions (analytical approach). In local-scale models, particle transport and deposition equations are solved by Computational Fluid and Particle Dynamics (CFDP) methods (numerical models). At present, analytical whole-lung models are the most appropriate deposition models for the study of health effects in the whole lung.

At the onset of inhalation, particles pass through the extrathoracic region, bronchial and alveolated airways, and after a short breath-hold time, follow the same

path back during the exhalation phase. Their depth of penetration into the lungs depends on the time during the inhalation phase at which they are inhaled. In terms of deposition calculations, this transport scenario requires the computation of deposition efficiencies in extrathoracic, cylindrical bronchial and alveolated airways, and quasi-hemispherical alveoli for the whole breathing cycle. The two primary differences between current models of inhaled particle deposition in the whole lung are the choice of the selected morphometric lung model, and the applied computational techniques, generally related to the complexity of the selected morphometric model. In general, five different classes of conceptual models with respect to lung morphometry and mathematical modeling technique have been recognized: (1) semiempirical regional compartment models (ICRP, 1994), (2) one-dimensional cross-section or 'trumpet' models (Taulbee and Yu, 1975), (3) deterministic symmetric generation or 'single/typical path' models (Yeh and Schum, 1980), (4) deterministic asymmetric generation or 'multiple path' models (Asgarian et al., 2001), and (5) stochastic asymmetric generation or 'multiple path' models (Koblinger and Hofmann, 1990). Models 2–5 are often termed 'mechanistic models', as they are based on a mechanistic understanding of physiological and physical mechanisms, while Model 1 is based primarily on mathematical fits to experimental data.

Indirect dose assessment approach

Several approaches exist for estimating human exposure. Direct methods involve exposure measures performed at the point of contact or uptake when the exposure occurs, while indirect methods involve extrapolating exposure estimates from other measures and existing data. A number of indirect exposure models (AERMOD, CALTOX, SHEDS, UKADMS, etc) are described by Fryer et al. (2006). In Buonanno et al. (2011, 2012b), activity pattern data were combined with microenvironmental data (human activities and PN size distributions) using an indirect Monte Carlo method simulation approach, to evaluate the doses of alveolar and tracheobronchial-deposited PN and surface area experienced by different age groups in Cassino (South Italy) and Brisbane (Australia). It was found that females received higher doses than males, as a result of their different lifestyle patterns, with females spending more time in indoor environments where higher exposure levels are experienced. With regard to age, adults (in particular, people aged 19–40) received considerably higher doses than teenagers (aged 15–18) and seniors (>65 years). This was also due to different lifestyle patterns, because the inhalation rates and other characteristics of the different age groups were found to have a negligible effect on dose. Overall, Italian daily alveolar PN and sur-

face area dose for all of the age groups was equal to 1.5×10^{11} particles and $2.5 \times 10^3 \text{ mm}^2$, while the alveolar PN and surface area dose received by all Australian age groups was equal to 3.0×10^{10} particles and $4.5 \times 10^2 \text{ mm}^2$, respectively.

The main reason for the significant impact of lifestyle on received particle doses is due to the different particle exposure levels experienced in different microenvironments. In particular, the highest dose intensities were found during cooking and transportation activities, including both indoor (car and bus) and outdoor (pedestrian and bike) means. Higher doses were received by Italian people compared with Australians, mainly because in Italy, particle concentration levels were significantly higher during eating, cooking, and transportation activities. When particle deposition as a function of the available air-tissue interface for different age groups is considered, infants and children (typically receiving a lower absolute dose) turned out to be exposed to higher normalized doses than those experienced by working adults.

Risk assessment of indoor aerosols

Previous chapters have illustrated published methods for monitoring personal exposure to indoor aerosols, their composition and the levels of human exposure, as well as the tools for dose assessment. They provide the information required for analyzing the potential impact that indoor aerosols may have on human health. While indoor epidemiology is challenged by difficulties in collecting exposure data from sufficiently large target populations, risk assessment can be conducted if an exposure-response relationship is available for the exposure metric selected for the risk assessment. General risk assessment methodologies are well established. However, there are specific challenges in applying these methods in the indoor environment, which makes the justification to give an overview of the general methodology in this particular context: our aim is to summarize how the methods have been specifically used for indoor exposures.

There are four major steps in risk assessment procedure for both carcinogens and noncarcinogens: (i) hazard identification, (ii) exposure assessment, (iii) dose-response assessment, and (iv) risk characterization (United States National Research Council, 1983; USEPA, 1992, 2005).

Hazard identification refers to identifying the pollutants of concern, and their potential adverse health effects, on the basis of the results of epidemiologic, clinical, toxicological, and environmental research. PM metrics and components identified as hazardous include $\text{PM}_{2.5}$ and PM_{10} mass concentrations, PN concentration (of ultrafine particles), PAH compounds, heavy metals (including Ni, Cd, Hg, Pb, V), elemental organic carbon, etc.

Exposure assessment quantifies the amount of the concerned pollutants that the receptor has been exposed to for a certain duration. The exposure value is then used to calculate the *intake dose*. It can be expressed in the terms of chronic daily intake (CDI, mg/kg day), given by:

$$\text{CDI} = \frac{\text{Average Exposure Conc.} \times \text{Inhalation Rate} \times \text{Uptake Fraction}}{\text{Body Weight}} \quad (3)$$

where the inhalation rate and body weight are commonly assumed to be $20 \text{ m}^3/\text{day}$ and 70 kg .

Dose-response assessment refers to estimating the probability and frequency illnesses occur. The potency factor is used as an indicator for the severity of adverse health effects. The values for commonly found pollutants can be found on the 'Integrated Risk Information System' (IRIS, USEPA, 1998). It can also be named the inhalation UR (UR) and can be expressed in terms of the slope factor (SF), where $\text{SF} = \text{Potency factors} \times (\text{body weight}/\text{inhalation rate})$.

Risk characterization combines the results from the dose-response and exposure assessments. The risk estimation methods for carcinogens and noncarcinogens are different. For carcinogens, with linear extrapolations, excess lifetime cancer risk can be calculated as:

$$\text{ELCR} = \text{SF} \times \text{CDI} \quad (4)$$

A commonly used acceptable value of ELCR is defined by USEPA as 1×10^{-6} (USEPA, 2005).

For noncarcinogens, with nonlinear extrapolations, risk can be expressed as a *hazard quotient* (HQ) (USEPA, 1992):

$$\text{HQ} = \frac{\text{CDI}}{\text{RfD}} \text{ or } \text{HQ} = \frac{\text{CDI}}{\text{RfC}} \quad (5)$$

where RfD and RfC are the inhalation chronic reference dose and inhalation chronic reference concentration, respectively. The acceptable level of HQ is 1.

These provide a general procedure for estimating the risks of exposure to any known pollutant, when the 'risk potential' (SF or dose-response) of the particular agent is known. For example, in research conducted by Guo et al. (2004), which estimated the ELCR of VOCs, the CDI was expressed by the authors as:

$$\text{CDI} = \frac{(\text{CA IR ED EF L})}{(\text{BW ATL NY})} \quad (6)$$

where CA is the mean contaminant concentration (mg/m^3); IR is the inhalation rate (m^3/h); ED, EF, and L are described as the total exposure duration in a year (hr); BW is the body weight (kg); ATL is the average lifespan (years); and NY is the number of days in a

year (365 days/year). CDI applies the unit of mg/kg/day. Potency factors are taken from the 'Integrated Risk Information System' (IRIS, USEPA, 1998). The standard values suggested by USEPA (1994) to assist in these calculations are as follows: (i) 20 m³ inhaled air per day, with an average body weight of 70 and 60 kg for adult men and women, respectively; (ii) 5 m³ inhaled air per day, with an average body weight of 10 kg for a child; and (iii) an average lifespan of 70 years.

The World Health Organization is using an environmental burden of disease approach and quantifying both carcinogenic and noncarcinogenic health risks using disability adjusted life years (DALY). This methodology was recently applied for a number of stressors, including ambient PM_{2.5} in European countries in the 'Environmental Burden of Disease in European Countries' (EBoDE) study (Hänninen et al., 2011). The approach was further specifically applied to indoor exposures of PM_{2.5}, mold particles and second-hand smoke (Hänninen et al., 2012). DALY is the measure of health impact due to a disease and is given by:

$$\text{DALY} = \text{YLL} + \text{YLD} \quad (7)$$

where YLL is the years of life loss due to premature mortality and YLD is the years of lost due to disability caused by morbidity. The primary model used in this study is a three-step process for obtaining DALY, as outlined below.

Relative risk identification means to provide a quantified level of risk due to the level of exposure:

$$\text{RR} = \exp\left(\frac{E}{E^\circ} \ln \text{RR}^\circ\right) \quad (8)$$

where RR[°] is the relative risk per unit exposure of E[°], and E is the exposure threshold.

Population attribution fraction (PAF) is the reduction in disease that would have occurred if the entire population was not exposed to the risk. This is given by:

$$\text{PAF} = \frac{f \times (\text{RR} - 1)}{f \times (\text{RR} - 1) + 1} \quad (9)$$

where f is the fraction of population exposed.

Environmental Burden of Disease is the final result which is given by:

$$\text{EBD} = \text{PAF} \times \text{BoD} \quad (10)$$

where BoD is the burden of the target disease in question, in years, given by the WHO. This gives the total DALY from a disease for the entire population, due to exposure to selected PM fraction. Currently, the

required RR parameters are well defined for PM_{2.5}, PM₁₀, and BC and are emerging for a number of other PM components.

In the EBoDE study, the burden of disease for PM_{2.5} included cardiopulmonary mortality, lung cancer mortality, total nonviolent mortality, chronic bronchitis, and restricted activity days. For PM₁₀, the diseases used for analysis included lower respiratory symptoms and new cases of chronic bronchitis. Results showed that the inhalation of suspended particulate matter posed the most severe health risk, with a DALY of 6000–10 000 per one million people. This was a much higher burden of disease when compared with other indoor exposures like second-hand smoke (600–1200) and radon (600–900).

These examples illustrate how some specific challenges can be handled for risk estimation in indoor environments. Also, exposure assessment in risk analysis is highly relevant and linked with the interpretation of E-R coefficients obtained in epidemiological settings. The net error in the assessment depends on the factor variability in the exposure measurement. Full handling of the complexity of exposure metrics and indicators would need to be linked with the personal exposure levels discussed in section 'Personal exposure levels' and the intake/uptake and deposition in the lung discussed in section 'Intake/uptake and deposition in the lungs'.

Studies on the risk assessment of indoor aerosols

This section reviews studies performed by researchers on the estimation of risks induced by carcinogens in an indoor environment. There were studies that complied with the given guidelines, as explained in the previous sections, and some which used modified methods for calculating the risks.

Studies following the guidelines. Studies that performed risk assessments, which complied with the given guidelines, included a study on the ELCR for various carcinogenic PAHs in Chinese, Malay and Indian food stalls (See et al., 2006). It used the same methods given by the USEPA, as illustrated by the various equations discussed above, for calculating exposure and dose for the PAHs contained in airborne particles. They reported that the ELCRs were 4.08×10^{-3} , 1.21×10^{-2} , and 1.07×10^{-3} for Chinese, Malay, and Indian cooking stalls, respectively, which is higher than the acceptable ELCR value provided by the USEPA.

It must be noted that cancer may also be caused by carcinogens in a gaseous form. The USEPA guidelines are comprehensive and are applicable in such cases. Studies using USEPA guidelines for gaseous carcinogens, including formaldehyde and acetaldehyde, have been conducted by Sousa et al. (2011), Cavalcante

et al. (2006), and Feng et al. (2006). They were performed in different environments in different countries. According to their findings, the cancer risks or ELCRs, ranged from the order of 10^{-8} in libraries in Brazil to 10^{-4} in hotel ballrooms in China.

Studies which modified the methods used in the guidelines. On the other hand, a few studies modified the methods used for exposure estimation and risk assessment, to adapt them to their assumptions and scenarios. See and Balasubramanian (2006) carried out a risk assessment for three metal noncarcinogens (Al, Cr, and Mn) and four metal carcinogens (As, Cd, Cr, and Ni) which were emitted by cooking activities in indoor environments. The physical and chemical properties of $PM_{2.5}$ were investigated for a Chinese food stall in Singapore that used gas stoves, and the average mass concentrations of metals were measured to estimate the risks. In addition to the four basic steps described in the guidelines above, the authors included a respiratory deposition factor to improve the accuracy of the exposure assessment. The deposition fractions of different sized PMs were considered with an interpolated equation:

$$E_i = -0.081 + 0.23 \ln(D_p)^2 + 0.23 \sqrt{D_p} \quad (11)$$

where E_i is the deposition fraction of particle size i , and D_p was the diameter of the particle.

Then, the overall deposition fraction (f) was calculated using:

$$f = \frac{\sum(F_i E_i)}{F} \quad (12)$$

where F and F_i are the number concentrations of all particles with an aerodynamic diameter $<2.5 \mu\text{m}$, and that of particle of size i , respectively.

Their results showed that the total HQ from noncarcinogenic metals is 1.54, which is approximately 50% higher than the acceptable value. The total ELCR from carcinogenic metals was 1.11×10^{-4} , which is about two orders of magnitude higher than the acceptable level proposed by the USEPA.

Studies on UFP risk assessment. Recently, researchers found that UFPs induce greater adverse health effects in humans than PM_{10} and $PM_{2.5}$ (Kennedy, 2007; Wang et al., 2009a; WHO, 2005). This drove an increase in the number of studies in this area, with a special branch of toxicology, nanotoxicology, also developed to assess health risks of workers in the nano-industry. In comparison, UFPs are much smaller than those which are the main contributors to PM_{10} and $PM_{2.5}$, and although prefiltration of inhaled UFP particles in the upstream nasal and bronchial airways can significantly reduce the deposition of particles

below about 10 nm in the peripheral alveolated airways (Hofmann, 2011), other particles in the ultrafine size range can cause more damage by penetrating deeper into the human respiratory system, resulting in possible inflammatory effects. UFPs also have a higher deposition rate, which causes an unknown degree of damage to humans (Araujo et al., 2008; Hirano, 2009; Oberdörster et al., 2005b). Similar to PM_{10} and $PM_{2.5}$, UFP-induced risks have been estimated based on quantitative data on dosage and response, however, unlike for the former, dose-response and exposure models dedicated to UFP are actively being developed but are yet to be standardized.

So far, researchers have attempted to use models similar to the guidelines for particle mass (PM_{10} and $PM_{2.5}$) given in WHO (2000) and USEPA (2005), with modifications to emphasize the deposition and penetration of UFP, to estimate the risks of inhalation of UFP. However, converting these guidelines to assess the risk of UFPs has proved to be a challenge. This is because researchers have concentrated on the link between the health risks and mass concentrations of PM; however, the mass concentration of UFP is insignificant compared with larger particles. In addition, even a small number of large particles would cause a large error in the measurement of UFP by mass. The key parameter that relates UFPs to health risks is thought to be their surface area, because UFPs have a higher surface area to weight ratio than larger particles. Indeed, the correlation of particle surface area to inflammatory response for nontoxic particles has been recognized by Oberdörster (1996) and Donaldson et al. (1998). However, different health effects aspects may be related to different dose metrics, such as size and number concentration (Grass et al., 2012; Oberdörster et al., 2005a). It is also conceivable that the inflammation response is triggered by individual particles depositing on the lung tissues.

Adverse health effects due to the inhalation of UFP are mainly found in the lungs, as the port of entry into the human body; however, the heart and brain have also been identified as important target organs. For example, there has been substantial research about the olfactory nerve as a pathway for brain exposure to ultrafine particles (Oberdörster et al., 2004). Translocation across the blood-brain barrier in certain regions of the brain may be another route of inhaled UFPs into the central nervous system, via the lungs and blood (Oberdörster et al., 2004). Possible illnesses due to the inhalation of UFPs include pulmonary fibrosis, pleural effusion, granuloma, and the increased risk of cancer. Similar to PM_{10} , the health risks are higher for susceptible people, including those with asthma or cardiovascular disease. There is also strong evidence supporting the link between UFPs and lung-related diseases. Researchers (Song et al., 2009) showed that factory workers who have been exposed to UFP for

5–13 months experience shortness of breath and pleural effusion. After a careful examination of the lung tissues, pulmonary fibrosis and pulmonary inflammation were diagnosed. However, factory conditions are unlikely to be the same as those in an ambient domestic environment, and therefore, these results are not directly useful for the public. In addition, it has been shown that for patients with cardiovascular disease, the inhalation of UFP from diesel engines poses a high risk of heart attacks (Miller et al., 2009). Although there are currently no studies proving a direct link between heart attacks and indoor UFP sources, it was found that people living close to traffic have a higher risk of suffering from coronary arteriosclerosis, which causes heart attacks (Hoffmann et al., 2007).

General cancer risk assessments involve analyzing the mass fraction of the chemical components of inhaled particles and calculating the individual cancer risks of the different chemical compounds, characterized by the potency factor. However, there are currently only a limited number of studies on the risk assessment of UFPs, resulting in a lack of data to construct the UFP equivalent of the potency factor. Some researchers endeavored to carry out a risk assessment on UFP by avoiding the dependency on potency factor using odds ratios to estimate the probability of cancer risk when exposed to UFP (Zalk et al., 2009; Zhao et al., 2006a,b). Another proposed method is to statistically estimate the probability of cancer based on exposure to different number and surface concentrations of UFPs by analyzing the number of affected patients (Chio and Liao, 2008; Liao et al., 2011).

Recently, a study by Sze-To et al. (2012) attempted to use the same approach used for PM to estimate the ELCR of UFPs generated and inhaled when cooking, based on the following equation:

$$\text{ELCR} = \frac{C \times \text{SF} \times Q}{\text{BW}} \quad (13)$$

where C is the exposure concentration, SF is the inhalation SF, Q is the daily inhalation rate (m^3/day) and BW is the body weight (and 70 kg is used). Furthermore, the equation was modified to suit the features of UFP. The modified ELCR (for the UFP-related part) was expressed as:

$$\text{ELCR} = \frac{\text{SF}_m Q}{\text{BW}} \left[C_f \int_{d_p} C_{\text{UFP}}(d_p) \beta(d_p) dd_p \right] \quad (14)$$

where SF_m is the SF of the UFP mixture, c_f is the conversion coefficient (in mg/nm^2) for diesel engine exhaust data (which was recently recognized as a carcinogen by the WHO (2012)), and C_{UFP} is the particle surface area concentration of UFP (nm^2/m^3). They reported that the average ELCR contributed by

UFP was 1.52×10^{-4} and 1.01×10^{-4} for cooking occupants and other occupants, respectively, which yielded results similar to some referenced epidemiological results (Wang et al., 2009b; Zhong et al., 1999).

Summary of the state of knowledge and recommendations for future research

This work was motivated by growing consideration of the scale, potential severity, and risks associated with human exposure to indoor PM. There have been several challenges identified, making the entire process of risk assessment, as well as its individual components, particularly complex. These include: (i) the vague understanding of the term *exposure*, as it is often adopted in indoor sciences; (ii) the intrinsic challenges of PM investigations and the still incomplete understanding of its overall impact on health; and (iii) the contribution from both indoor and outdoor sources to exposures occurring indoors. Indoor exposure to PM is often divided into two components, differing significantly in composition, temporal patterns, and their relationship to personal time–activity patterns: (I) particles from outdoors, which can significantly affect PM levels in indoor and traffic related microenvironments; and (II) particles from indoor sources, which have the potential to generate very high levels of PM that do not correlate with outdoor levels on a temporal basis. Health responses associated with PM originate mostly from ambient epidemiological studies, which are not suitable for evaluating the composition-specific toxicity of indoor-generated particles. However, in studies attempting to identify the difference constituents in ambient PM, the results are very vague and heterogeneous, suggesting that particle mass, and not composition, may be the best indicator of PM toxicity. If this indirect finding is expanded to indoor-generated particles, partly supported by their similar composition, indoor-generated exposures should be treated just as seriously as exposure to ambient PM.

This review shows that personal exposure to indoor particulate matter has traditionally been determined from measurements carried out at fixed locations, in ambient outdoor or indoor microenvironments (see section ‘Methods for monitoring personal exposure to indoor particulate matter’). Even if such estimates are commonly used, these are not as reliable or as accurate as monitoring by personal samplers that are worn or carried by the subjects. In this article, we have provided a general summary of the various methods and instruments which have been used for the characterization of PM in indoor microenvironments, for the purpose of measuring personal exposure. With the advances in technology, particularly miniaturization, there has been a rapid proliferation of compact instruments that can be used for personal monitoring. While hand-held

instruments for monitoring gases, such as carbon dioxide and carbon monoxide, have been around for many years, these are now being matched in size by instruments that can measure particle mass, number, and surface area. In the near future, we can no doubt expect to see instruments that measure gases, such as nitrogen dioxides, and a range of other chemical species.

There is a growing body of literature available on exposure to indoor aerosols in the residential environment. Even so, not all of the results from these studies can be directly compared, due to the differences in study design and instrumentation; however, a general picture of the magnitudes and trends in exposure is starting to emerge (as discussed in section 'Residential environment'). In particular, it has been shown that, in limited studies assessing daily integrated residential exposure to UFPs, the contribution of indoor sources was ~50% (ranging from 19% to 76%). This not only indicates a strong dependence on resident activities, source events and site specificity, but it also highlights the importance of indoor sources for total personal exposure. Median values of reported indoor PM_{10} concentrations have been shown to be slightly higher than outdoor concentrations, while median indoor and outdoor values for $PM_{2.5}$ were similar, but with higher variations seen indoors. In addition, the median value $PM_{2.5}$ concentrations from personal monitoring is also higher than indoor and outdoor concentrations. On the basis of the reviewed articles, a general need is evident for: (i) characterization of indoor residential sources by means of particle chemical composition, toxicity, size-dependant physical properties affecting exposure and dose characteristics; (ii) personal exposure monitoring with portable devices for better characterization of exposure (allowing for the capture of near-field exposures that are not well characterized by stationary monitoring); (iii) apportionment of the contribution of indoor residential and outdoor particles to personal exposure and their quantitative assessment on population representative basis; (iv) modification of outdoor particles by buildings and the effect on exposure (composition, size, dose, etc); (v) use of a normalized metric for integrated personal exposure assessment to enable meaningful comparisons (concentration h/day); (vi) characterization of microenvironmental concentrations for times when the person in question is within a given space (e.g., residents present at home); and (vii) international effort to standardize (possibly through ISO) the requirements for comparable experimental studies.

A review of exposures in the school environment (see section 'Schools') concluded that, in classrooms, PM_{10} concentrations during school hours were higher than outdoor concentrations, whereas UFPs significantly increased with increasing truck traffic density

and significantly decreased with increasing distance from the road. It was also concluded that, in relation to school environment, the scientific literature is largely insufficient with respect to: (i) monitoring personal exposure to particles in schools; (ii) UFP monitoring (limited to a few papers); (iii) concentrations based on an averaging time that only captures when the person in question is present in a given space (i.e., children are at school); (iv) the relationship between ambient concentration and personal exposure; and (v) the contribution of school exposure to a child's daily exposure, with respect to other sources (transportation, home, outdoors, etc).

It was shown that many different processes influence the chemical composition of indoor PM, and the size-resolved composition may change dynamically as certain sources or losses become more or less important over time. However, due to instrumental limitations, exposure studies have only analyzed integrated filter samples to date. The next generation of exposure studies should seek to improve this limitation, either by pairing gravimetric samples with time-resolved indoor aerosol measurements (such as with an aerosol mass spectrometer) or with new instruments developed specifically for this purpose. Personal exposure sampling for microbial aerosols is an emerging technology, and it has only been used in a handful studies to date. Most previous work has focused on the composition of biological particles in a given microenvironment, based on fungal or bacterial spore/cell counts. As a result, there is very limited exposure data available for bioaerosols, from a limited number of countries and for a limited number of microenvironments.

Moving on to the next element of risk assessment, deposited dose, current whole-lung deposition models permit the prediction of particle deposition in a single airway or in airway generations, for any combination of particle size and breathing pattern, except for the semiempirical models, which are restricted to regions accessible to measurements. By integration over a defined sequence of airway generations, average generational, lobar, regional, or total deposition fractions can be obtained. As all presently available whole-lung models have been validated by comparison with experimental data on total deposition, they represent versatile and reliable computational tools for linking personal exposure with the deposited dose and thus, for use in the risk assessment process.

A different approach to dose assessment utilized indirect Monte Carlo method simulations to evaluate the doses of alveolar and tracheobronchial-deposited PN and surface area experienced by different age groups. The application of this method for Cassino (Southern Italy) and Brisbane (Australia) populations highlighted that, due to different lifestyle patterns, there was a difference in dose received by gender (females higher than males), age [adults (aged 19–40)

higher than teenagers (aged 15–18) and seniors (>65 years)], and country of residence (Italians higher than Australians).

Risk assessment methods for analyzing the health impacts due to exposure to indoor aerosols were reviewed, based on the standard guidelines for particles measured in terms of mass concentration (PM₁₀ and PM_{2.5}). These guidelines included four major steps: hazard identification, dose-response assessment, exposure assessment, and risk characterization. For cancer risk quantification, excess life cancer risk (ELCR) was calculated for a given dose and potency factor for the carcinogens of interest, with most studies that followed these guidelines yielding valid results. DALY were used to indicate the environmental burden of disease caused by PM and its effect on the population. It includes analysis of the adverse health effects other than cancer and results showed that the inhalation of PM was a significant factor affecting public health. A few modifications were developed by researchers to emphasize the effect of deposition of different sized particles. This was especially necessary in UFP risk assessments, because the damage to humans from UFP inhalation are mainly due to their surface area and number, which were overlooked in current guidelines that focus on mass concentration. For example, the results from dose-response assessments, which are based on the mass concentration of chemicals, are unsuitable for UFP risk assessment. Therefore, more data on the damage caused by surface area and number concentration of UFPs are needed and should be compiled from toxicological and epidemiological research. A more sophisticated and quantitative theory on how UFPs cause damage to humans would help to produce a set of suitable guidelines for risk assessment.

While the total burden of disease associated with PM has been estimated to be as high as 6000–9000 DALY per million inhabitants (Hänninen et al., 2011), the potential contribution of indoor-generated particles is higher than most of the other environmental pollutants. Crude estimates, based on exposure partitioning, suggest that the burden of disease from indoor-generated particles could reach the order of 1×10^3 – 3×10^3 healthy life years lost per million inhabitants in developed countries every year. This represents 10–30% of the total burden of disease from PM exposure. In developing countries, poor indoor air quality is one

of the leading causes of poor health, especially for mothers and young children, who spend most of their time at home. Therefore, exposure to PM caused by indoor sources is likely to be one of the dominant environmental factors affecting human health globally. Due to the challenges associated with conducting epidemiological assessments, the role of indoor-generated particles has not been fully acknowledged, but fortunately, the situation is beginning to change. Improved exposure and risk assessment methods are needed, together with a serious focus on exposure control. An example of this could be investigations aimed at directly linking exposure with response (*exposure–response*) or with individual risk, or assessing the impact of proposed regulations or policy measures on actual reductions in exposure (Morawska et al., 2011b).

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Table S1 Summary of the existing methods which have been used for area and micro-environment monitoring of PM.

Table S2 Summary of studies included in this review assessing indoor (in residences) and outdoor concentrations, as well as concentrations from personal monitoring and integrated daily residential exposure (note that ‘concentrations from personal monitoring’ were presented as ‘personal exposure concentrations’ in the original articles, but given that exposure is a product of concentration and exposure duration, this has been changed to ensure consistency within this review).

Table S3 Traffic-related air pollutant concentration values in schools reported in different studies.

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