

REVIEW

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Fine particulate matter in indoor cultural heritage: a literature review

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Abstract

Fine particulate matter is, on account of its aerodynamic properties and typical composition (especially diesel particulate matter and carbonaceous particles) the particulate pollutant potentially most harmful to cultural heritage, representing an aesthetic issue and an agent of chemical degradation simultaneously. This paper reviews the current knowledge of the life-cycle of fine particulates, focussing on diesel particulate matter from emission to deposition, including its aesthetic and chemical consequences, and draws attention to some imbalances in the current state of research. The currently available measurements are biased towards coarse dust, and information on the consequences of particle deposition is largely restricted to the outdoor environment. More evidence on the chemical effects of the most common types of fine particulate matter in typical indoor materials is needed to enable risk assessment for indoor collections.

Keywords: Fine particulate matter, Diesel particulate matter, Soiling, Deposition, Resuspension, Indoor air quality, Indoor heritage

Introduction

Most museums, galleries, libraries and archives, as well as many historical palaces and houses, are located in urban centres, surrounded by a complex and changeable urban atmosphere. While the last twenty years have seen a great reduction in the emissions of the pollutants that have been typically considered more harmful for vulnerable heritage materials, scientific interest is shifting towards other pollutants and particulate matter (PM) has been a subject of great interest.

However, PM can be an elusive subject of study. Different sizes display different properties, typical sources and even different behaviours. Studies of PM in indoor heritage environments have generally focused on dust, which is one of the characteristic sizes -the largest- of PM. This bias towards coarse particles is evident if we look at the minimum diameter of the particles collected in different monitoring campaigns in the field of heritage science. A survey of 25 scientific papers [1-25], most of them reviewed here, with the keywords “heritage” and “particulate matter” reveal that 32% of them analyse only particles

up to 10 μm and 16% include particles up to 2.5 μm , i.e. more than half of the studies did not look into submicron particles. If a study did take into account particles $\sim 0.5 - 1 \mu\text{m}$, it is generally the lowest size mode considered (36% of cases) and no particles are studied under this value. Finally, in 92% of the cases, particles are studied in only two or one size modes (usually 2.5 and 10 μm). However, two size fractions are not enough to reflect the actual size distribution, which is only analysed in a minority (8%) of the studies.

Obtaining size distributions and specifically quantifying the amount of submicron particles is common practice in aerosol monitoring outside the heritage field. All these sizes, and not only large particles, have a certain role in indoor heritage environments. In fact, coarse particles exhibit characteristics of great interest to conservators: they are significant carriers of mass to surfaces, and, being bigger, are more likely to alter the visual appearance of objects. But their number concentrations are orders of magnitude smaller than the concentrations of fine particles, especially in urban environments. Additionally, the composition of coarse and ultrafine particles is also different, and while small particles might carry less mass, they may carry components of different reactivity.

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This review deals with fine particles, i.e. all particles smaller than $1\ \mu\text{m}$, without excluding particles smaller than $0.1\ \mu\text{m}$, commonly referred to as ultrafine (UFP). This size range is clearly separated from coarse dust, and it includes pollutants of particular interest to conservation of cultural heritage. Special attention will be given to particles derived from combustion present in urban environments, particularly Diesel Particulate Matter (DPM).

The review serves a double purpose. First of all, it attempts to identify if fine and ultrafine particles can be regarded as a relevant risk to cultural heritage indoors. It also identifies multiple areas that require further research. Secondly, it aims to provide a guide to heritage managers and curators interested in the properties of this particular pollutant. While aerosols indoors have been studied in detail, no account exists of the distinct behaviour of fine particles. Since not many investigations deal directly with fine and ultrafine particles in heritage environments, this review will make use of work produced in other fields -aerosol science, environmental science, computational modelling- which can be applied to heritage at least partly.

Sources, trends, and projections

The term PM identifies all the particles that can be found in the atmosphere, in other words, those that can be suspended in air and transported by it before they deposit. This includes particles composed only of several molecules, with diameters around $0.01\ \mu\text{m}$, up to coarse dust with diameters around $100\ \mu\text{m}$. Samples of atmospheric PM usually display a very characteristic size distribution, clearly separated in three different size modes (Figure 1) [26]. These modes are referred to as the nucleation, ultrafine, or Aitken mode ($<100\ \text{nm}$), coagulation

or accumulation mode ($100 - 1000\ \text{nm}$) and coarse mode ($>1\ \mu\text{m}$). An alternative and common nomenclature is using PM_x for all the particles smaller than x in μm , the usual values being 10 , $2.5\ \mu\text{m}$ or, less commonly, 5 and $1\ \mu\text{m}$. Although the use of this notation is widely spread, it makes an artificial division between the actual size modes. E.g., $PM_{2.5}$ is in the middle of the coarse mode but is generally taken as a good indication of the amount of anthropogenic particulate pollutants in urban environments. This nomenclature is useful from the health perspective as it approximately denotes the fractions which can penetrate to different depths of the human respiratory system. It is not, as we shall see, particularly suited for heritage purposes. However, its status of a standard makes its use unavoidable.

Given such a broad definition, it is natural that there are a variety of origins, sources and compositions of PM. Nonetheless, there are a limited number of relevant sources. The majority are related to energy production, and natural sources (such as sea-spray) are often negligible in comparison with the anthropogenic ones. Among them, the combustion of fossil fuels, especially in road transportation, has a prominent role.

In the UK, road transport is responsible for more than half of the particles of the smaller size ranges and around 20% of the larger [27]. This relative contribution is even greater in urban areas. At Marylebone Road, arguably one of the most polluted streets in London, traffic-generated particles make up to 27% of the PM_{10} mass concentration and 42% of the $PM_{2.5}$ [28]. Most road transport particles are due to diesel vehicles (e.g. 72% of USA road transport PM) [29]. Even though this picture varies greatly between regions, similar particle source apportionments have been reported by different studies carried out elsewhere [30,31].

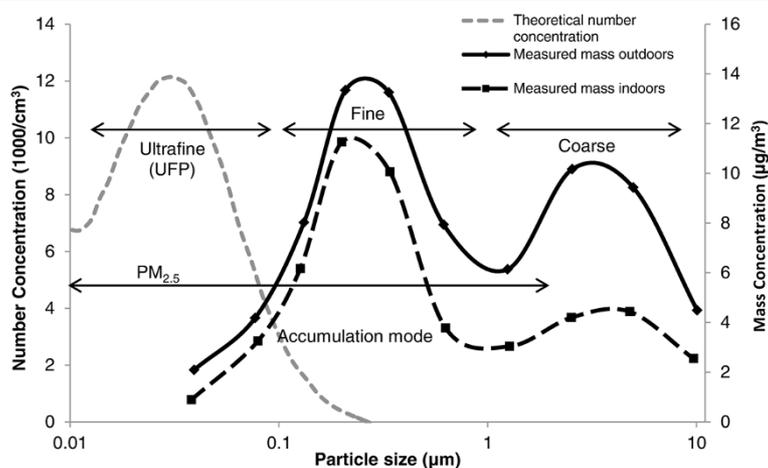


Figure 1 Typical size distribution. An example of distribution of atmospheric particles showing the relevant and typical properties of DPM. Mass concentrations adapted from measurements taken in the National Library in Prague [24] and completed with theoretical number concentration of ultrafine particles according to [26].

Emissions of all types of PM are predicted to decrease in the decades to come, including emissions of the smallest particles [32] due to the implementation of mitigation strategies such as diesel soot filters and the substitution of fossil fuels with alternative energy sources. Analysis of global emission trends under different scenarios reveal that emissions of traffic-generated PM will reduce in all the continents except in Africa, where they are predicted to increase 1.3 - 3.1% per year, depending on the scenario, due to economic development and ageing car fleet. Global emissions from vehicles will reduce 1.3 - 2% on average per year in the following 40 years [33]. China will achieve a reduction in emissions of black carbon of 9% by 2020 [34].

The PM concentration limit suggested by the World Health Organisation (WHO) [35] and the European Commission [36] for $PM_{2.5}$ is $25 \mu\text{g}/\text{m}^3$, and the US Environmental Protection Agency has suggested a value of $13 \mu\text{g}/\text{m}^3$. Even though these limits are exceeded in some regions, it is likely that they will be satisfied in the near future. However, WHO states as no threshold for PM has been identified below which no damage to health is observed, the recommended value should represent an acceptable and achievable objective to minimize health effects in the context of local constraints, capabilities and public health priorities [35]. The same logic may be applicable to heritage materials. It can be concluded that, even though emissions are gradually decreasing, traffic-generated pollutants will remain an important part of urban atmospheres for decades, especially in developing economies.

Particles derived from combustion

The laser-cleaning literature abounds with uses of the word “soot”, referring to dark deposits on indoor and outdoor materials. However, no standard description or characterisation of soot exists. The concept of “black

carbon”, extensively used in aerosol and environmental science, suffers from a similar imprecision, sometimes used interchangeably with soot [37].

What is clear is that fine particles (PM_1) mostly originate from combustion processes. Morphology and composition of these particles varies with the source to some degree: burning of candles, coal, tobacco or diesel fuel. These particles also have common features: a high content of inorganic carbon that will display high light absorbance, combined or coated with traces of other elements, and an aggregate-like shape.

Diesel particulates

A typical diesel particulate is shown in its graphic representation in Figure 2a and a TEM micrograph in Figure 2b [38]. Diesel particulates in the accumulation mode have a distinct morphology consisting of a self-similar agglomeration of primary particles, which has led some researchers to use fractality as a characterisation factor [39]. These primary particles are mainly composed of elemental carbon with metallic traces, and are coated with a layer of organic carbon and sulfate. This composition is subject to some variability as it depends on the engine type, the engine load and the fuel used. Total carbon usually accounts for 75 - 90% of the overall mass [40], and the metal traces can include, in the order of relative abundance: Ca, Fe, Mg, Zn, Cr, Ni, Ba, Pb [41,42]. Elemental carbon (EC) is a characteristic component of DPM, which is typically the source of 90% of the EC in urban environments [43]. Organic Carbon (OC) may be emitted directly into the atmosphere or can be formed in gas-to-particle atmospheric processes (secondary aerosol). EC, on the other hand, emerges primarily from the combustion of carbonaceous matter, and its presence is generally regarded as a good indicator of the fossil fuel combustion origin of PM, especially in urban areas. Recently,

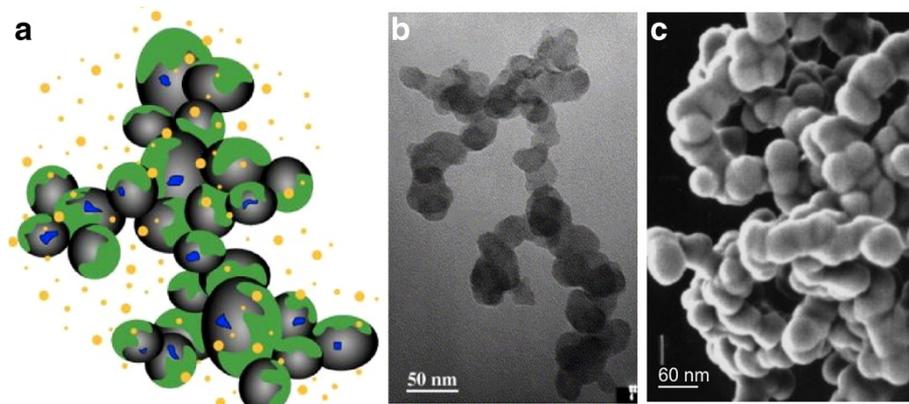


Figure 2 Combustion-derived particles. **a)** Graphic representation of a typical diesel particle containing all its components: EC substrate, OC coating, metal traces and nucleation particles, taken from [41]. **b)** TEM image of a diesel exhaust particle with a magnification of 230000x, taken from [38]. **c)** A candle soot particulate displaying a morphology similar to that of DPM [46].

however, it has been claimed that atmospheric EC can originate from sources other than diesel exhausts [31,44], which makes source apportionment difficult. Polycyclic aromatic hydrocarbons (PAHs) and *n*-alkanes have also been suggested as possible tracers for different particulate pollutants generated by road traffic [45].

Other combustion particulates

Even though this review focuses on particles of outdoor origin, mention should be made of other specific sources of fine and ultrafine particles which may be of interest. Candle-burning soot, for example, is commonly associated with indoor deposits in temples and churches. It has been found that the amount and composition of particles emitted from candles depend on the burning mode. If the flame is in steady state, it emits a relatively high number of ultrafine particles dominated by either phosphates or alkali nitrates originated from additives. Sooting burn, in addition, emits larger particles mainly consisting of agglomerated elemental carbon, with a morphology which is similar to DPM [47] (Figure 2c). Particles with the same morphology [46] and similar composition can be emitted during cooking [48], an activity not unusual in large heritage sites and museums. A “black deposit” or “soot deposit”, must, therefore, be assessed with care in order to identify the most likely origin of the particles.

Concentration trends indoors

The indoor PM concentration is generally a reflection of the outdoor concentration. Certain indoor activities represent exceptions to this rule. It has long been established that different activities, such as cooking, housework, or simply any physical activity, result in concentration peaks over the baseline set by the outdoor concentration [49].

The frequency of these activities in heritage environments, and the efficiency of air cleaning systems define the daily PM pattern.

Fine PM such as DPM originate from outdoor sources, and the events that can cause a drop or a sudden increase in its concentration are rare. Figure 3 shows the variation of fine particle ($d < 1\mu m$) concentration in a roadside house. It can be clearly appreciated that the indoor concentration is a consequence of the outdoor concentration, except when particles are emitted during cooking activities.

In heritage environments, indoor events that lead to emission of fine particles are uncommon. Actions such as cleaning, or physical activity cause variations in the concentration of the larger particles, but generally leave the smaller size modes unaltered. Figure 4 shows the variation of airborne particle concentration in the Correr Museum, Italy [17]. It can be appreciated that the peaks of 10, 5, 2, and 1 μm particles, which correspond with the opening hours of the museum, as well as their high variability, are not reflected in the concentration pattern of the finer particles, which varies smoothly. Very similar temporal evolutions have been reported in the Anatomy Theatre of Padua [7] and in the Chiericati Municipal Museum of Vicenza, Italy [50]. As a general rule, while physical movement increases the number of coarse particles, actions involving heating tend to increase the concentration of the smallest particles. Air heaters, electrical radiators and stoves have been found to increase concentrations of particles between 0.02 - 0.1 μm up to 100,000 - 200,000 $particles/cm^3$, while leaving larger particles unaltered [51].

The mass and number concentrations seen in Figures 3 and 4 are representative of the typical concentrations in indoor environments. The average concentration of PM_{10}

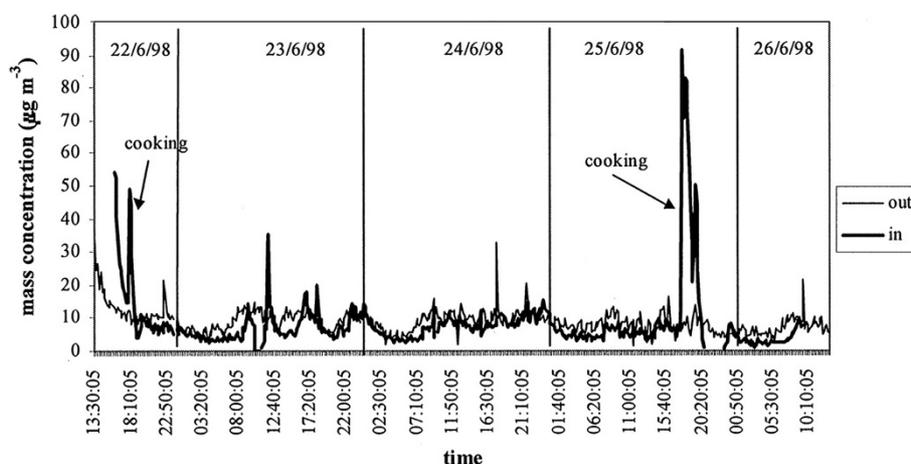


Figure 3 Outdoor and indoor variations of PM concentration. Indoor and outdoor variations of fine particle concentration ($d < 1\mu m$) in a roadside house in Birmingham (Taken from [49]).

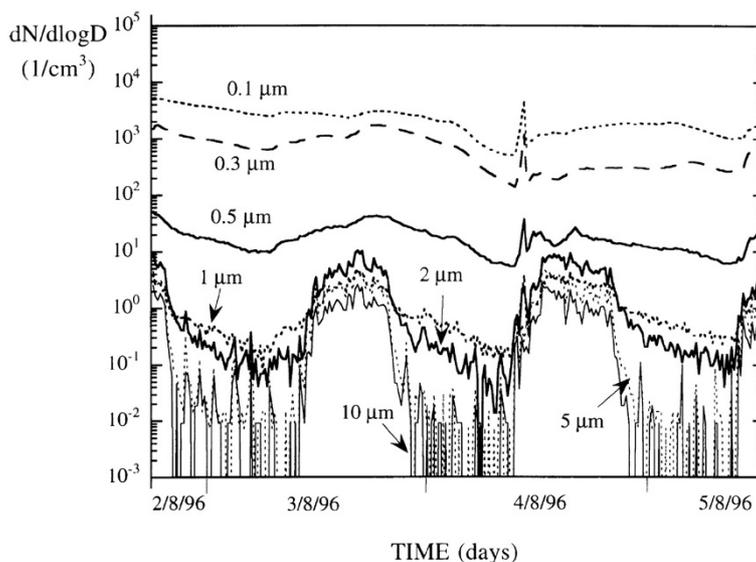


Figure 4 PM in the Correr Museum. Variation of particulate matter concentrations in the Correr Museum, Venice, Italy, taken from [17].

inside the Alhambra, Granada, Spain, was $17 \mu\text{g}/\text{m}^3$ in summer and $8 \mu\text{g}/\text{m}^3$ in winter [2], and its most abundant component was black carbon. Traffic was found to be the major source of fine particles. Between 10 and $20 \mu\text{g}/\text{m}^3$ of $\text{PM}_{2.5}$ were detected in display rooms in the Plantin-Moretus museum in Belgium [19], and $40 \mu\text{g}/\text{m}^3$ in the Archaeological Museum of Thessaloniki, Greece [12]. If total suspended particles are measured, higher values should be expected, e.g. $60 - 70 \mu\text{g}/\text{m}^3$ inside the Wawel Castle Museum in Cracow, Poland [13]. At this site, particles containing elemental and organic carbon were found to be the ones that penetrate more easily into the museum. Even though particle concentrations in heritage locations are commonly measured in this manner, they tell us little about the fraction of fine particles that penetrate into these locations. Analysis of the bulk chemical composition of the collected particles is a common further step, but it is rare to find more detailed measurements of size distribution including fine particles. A good example are the detailed measurements taken in the Czech National Library in Prague [24], or in some Californian museums [25] which display a clear bimodal size distribution (Figure 1).

The indoor/outdoor ratio (I/O ratio) is widely used to describe the differences between the indoor and the outdoor environments [52-55]. There is a great variability in the measured PM I/O ratios in particular cases, but in general, some rules of thumb can be extracted from experience. Firstly, as shown in Figure 5, the I/O will be higher for particles of size between $0.1 - 1 \mu\text{m}$. An explanation for this phenomenon is given by the aerodynamics of these particles, and will be further explored in the next sections. Secondly, I/O ratios for particles containing EC (which in

urban atmospheres can be understood as DPM), the I/O ratio are typically $0.5 - 0.9$ [49,52,56].

The life-cycle of fine particles indoors

Fine particulates penetrate into buildings not only through large openings but also through cracks and filter inlets. At the end of its indoor life, PM deposits on surfaces or is removed by mechanical or natural ventilation. Some particles, especially the larger ones, may

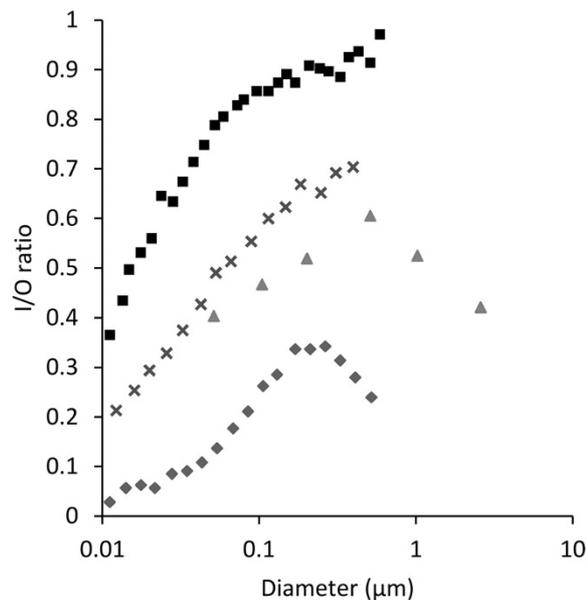


Figure 5 I/O ratios. Experimental I/O ratios as a function of particle size. Values taken from [53] (diamond), [54] (square), [55] (triangle), [52] (cross).

be re-suspended and re-deposited. This set of inlets and outlets of PM summarizes all the steps that ultimately determine the extent of soiling of indoor surfaces. They have been extensively studied as separate phenomena, and considerable literature exists describing each process experimentally, or with empirical or semi-empirical correlations. The work of Nazaroff [57] is the only piece of work presenting a comprehensive summary of all the possible particle flows in any given building. There are other published balances of PM indoors [58], but generally, as they focus mainly on practical health issues, deposition is not studied nor are the minor penetration processes, such as leakage. Also, while health scientists are concerned with the free volume, heritage scientists should be concerned with the room as a whole, and consider both the volume and the surfaces. Figure 6 summarizes, in an approach similar to Nazaroff's, the main particulate matter fluxes in a room.

Penetration

First and foremost, particles enter through main inlets: windows, doors, and any openings that connect the indoor and outdoor environments, such as gaps under doors. The use of windows for ventilation can quickly increase the particle concentration indoors to match the outdoor levels [59] for all particle sizes. For example, a study of PM in Californian museums [60], such as the Sepulveda house, which lacks an air filtration system,

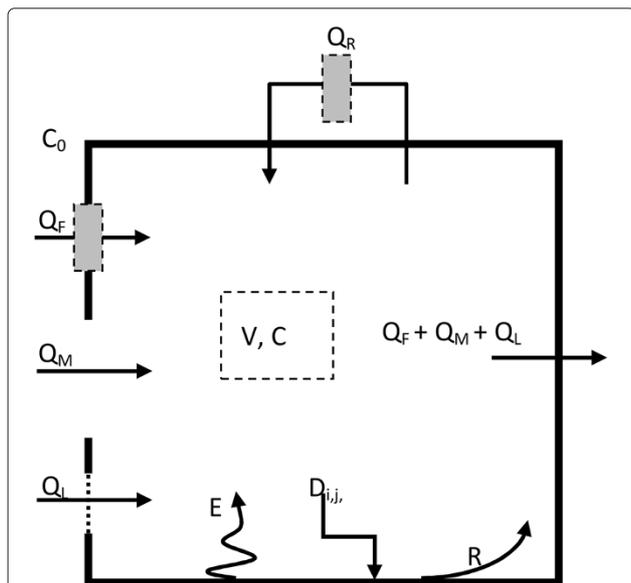


Figure 6 Summary of particle fluxes indoors. The possible inlets and outlets of particles. Q_M is a main inlet, such as an open window, Q_L the leakage, Q_F the filtered inlet, Q_R an internal filtered recirculation, E the internal emission, R the re-suspension, D_{ijk} the deposition to walls, floor, and ceiling, C_{n0} and C_n the outdoor and indoor concentrations of every particle size fraction, and V the room volume. Both Q_F and Q_R are subject to certain efficiency.

showed I/O rates equal to 1. This implied that the risk of deposition indoors was equal to the one outdoors. Leakage, or the penetration of particles through cracks or gaps in building envelopes, has also been extensively studied both experimentally and theoretically. It is usually a major particle source in buildings that rely solely on mechanical ventilation [61]. The fundamental difference between a crack and a large opening is that a significant fraction of particles will deposit on the internal surfaces of the crack, and therefore leakage has a certain penetration efficiency which depends on particle size. Ultrafine particles led by Brownian motion will tend to deposit on crack walls, and coarse particles will also deposit quickly on upward facing surfaces [57]. Only the accumulation mode (0.1 - 1 μm) will cross the crack efficiently. Liu and Nazaroff [62] developed a simple mathematical model to estimate the penetration efficiency of particles, and it has been successfully used in several experimental studies since then [63,64]. Figure 7 shows a solution of the Nazaroff equation. Note that the particles between 0.1 - 1 μm penetrate with an almost 100-% efficiency through the crack. Although generally regarded as a minor source, crack infiltration has been found to account for 73% of the PM_{10} indoors in a poorly ventilated building [65]. Leakage can lead particles to the interior of showcases. The comparison of particle size distribution inside and outside a display case in a museum in Padova, Italy, revealed that the I/O ratio was 1 for all particle sizes, except for particles larger than 1 μm [66], which is in accordance with the theory.

Indoor emission

Indoor sources of fine particles are found in some specific heritage environments, such as in-use churches where

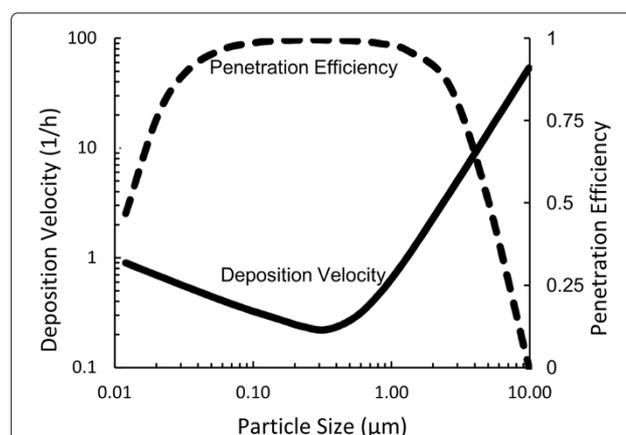


Figure 7 Leakage through cracks. Comparison of deposition and penetration through cracks and leaks obtained with the Nazaroff equations for deposition and leakage. Note that the particles with smaller deposition rates also penetrate through cracks more efficiently. Crack dimensions: width = 0.20 mm, length = 4 cm, pressure difference = 4 Pa, air flow conditions: $K = 0.5$, cubic room (3x3x3 m). All values taken as representative of realistic cracks by [62].

incense and candles are burned. An increase for a factor of 9.1 in the concentration of PM_{10} has been found after services that involved incense burning in Ruhr, Germany. In this case, the concentration inside the church remained above the outdoor levels for ~ 24 h approximately [67]. These findings are consistent with values found during services in medieval churches in Cyprus, where indoor $PM_{0.5-1}$ concentration was found to be up to 10.7 times larger than the outdoor concentration.

A relatively unknown indoor source is the thermal desorption of organic compounds and emission of submicron particles from household dust [68,69]. At temperatures above 50 °C, which are often present in indoor environments, concentrations around 2500 *particles/cm*³ can be generated [69]. Investigation of this phenomenon, which to the best of our knowledge has not been researched in heritage sites, should be considered when introducing new heating points in the environment.

Movement

Air transport

The airflow in an indoor space, isolated from the outdoor climate, is governed predominantly by two factors: temperature gradients and mechanical mixing. In a totally isolated room, only the temperature profile will define the air velocity pattern. Studies of indoor micro-climates show that temperature gradients are a consequence of a number of factors such as presence of heating points, proximity to windows, temperature of the surrounding spaces, human presence, lighting or similar. Heat sources induce vertical convective flows that displace contaminants upwards in an enclosed space. Cool vertical surfaces, such as windows, induce downward flows, which results in a circular movement of air around the room. A typically observed air movement pattern is the upward flow from radiators or air circulation behind furniture or paintings due to the difference of temperature between the wall and the air [70]. PM is largely transported by the movement of the surrounding air. In other words, the Stokes number, the non-dimensional parameter which describes the behaviour of particles in suspension, is generally well below 1 ($St \ll 1$). The Stokes number, $St = \tau U/D$, is determined by the ratio of the relaxation time of the particle (τ), the characteristic dimension of the obstacle obstructing the fluid flow (D) and the velocity of the fluid (U).

Particles with $St > 1$ will have their own velocity field and enough inertia to detach from air streamlines, and particles with $St < 1$ follow the air current closely [71]. However, the velocity field of particles does not coincide completely with the velocity field of air. Particles have a certain mass, and therefore their velocity has a vertical component due to gravitational settling. Coarse particles settle down gravitationally much faster than fine particles, and this creates a certain stratification of the

concentration. Measurements in indoor domestic environments have revealed a higher proportion of $PM_{2.5}$ on the upper parts of rooms, and higher abundance of PM_{10} towards the floor [51]. Measurements of particle deposition in ceiling, walls and floor reveal that almost no coarse particles deposit on the ceiling, while all the deposited mass in the floor is due to coarse particles [25].

Other transport mechanisms

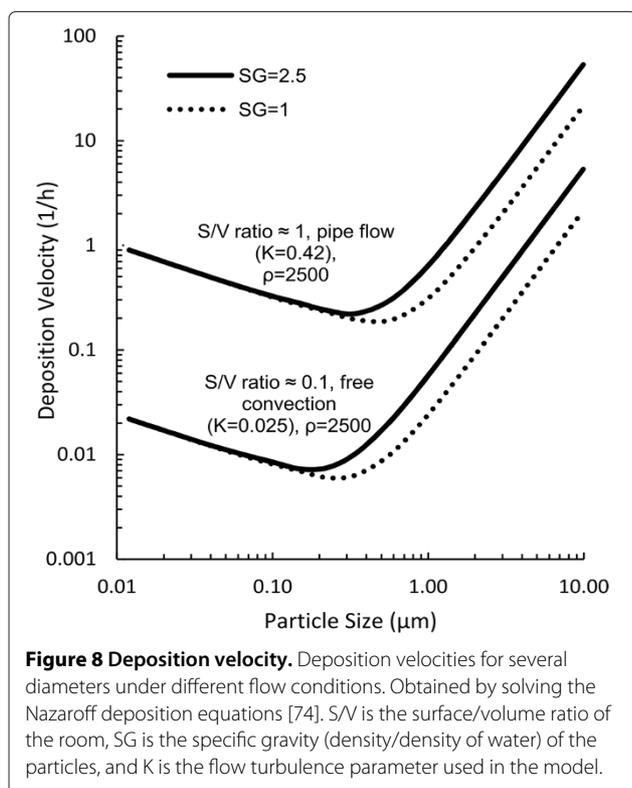
The smallest particles are largely affected by Brownian diffusion (also called “random walk”), which is a result of collisions between particles and air molecules and occurs in all directions. In any given room, coarse particles will be found in areas with the highest air flow, while fine particles will tend to diffuse around all the available space. Thermophoresis, the displacement of particles from high to low temperatures, is a phenomenon also common indoors. The balance between air transport, diffusion and thermophoresis has been studied in detail by Camuffo [72] in the case of the soiling of murals. It was pointed out that when a vertical fresco is colder than the surrounding air, the temperature gradient forces thermophoresis towards the wall, and at the same time a downward free-convection flow develops, resulting in an overall increase of deposition rates. When a fresco is warmer than the air thermophoresis takes fine particles away from the wall, but this effect may be counteracted by an upwards convective flow that increases deposition of coarse particles. The best situation for conservation purposes is, therefore, a thermal equilibrium between wall and air, whereas cold walls are the less desirable scenario.

Deposition

The deposition velocity of PM varies with particle diameter. This dependence is well known, and several authors have suggested mathematical expressions to estimate deposition velocities under different conditions. Perhaps the most used is the model derived by Nazaroff et al. [73,74], which has been successfully applied to museum environments [75]. Figure 8 has been obtained by solving this model, and shows the range of values of deposition velocity that can be expected under different flow conditions and for different diameters.

Dependence on particle diameter

The highest deposition rates are found for the largest particles (1 - 10 μm), which are governed mostly by gravity and tend to deposit on horizontal surfaces, and for the smallest particles (0.01 - 0.1 μm), which are mostly governed by Brownian motion and tend to diffuse and collide against floor, walls or ceiling. The mass of particles is only relevant in the larger size fraction, in which larger densities mean larger deposition velocities. Between these two size modes, the accumulation mode (0.1 - 1 μm) shows the



slower deposition rates, which are up to 2 orders of magnitude smaller than that of the coarse particles [57]. These lower deposition rates imply that particles in the accumulation mode tend to remain in suspension for longer, and therefore travel longer distances. In other words, while the coarse particles will deposit shortly after penetration indoors, near the source, the accumulation particles will distribute more evenly around the available space [76].

Dependence on air flow

The flow turbulence parameter, K , a key component of the Nazaroff deposition model, represents the turbulence regime of the air. It is an influential parameter and at low values of K , when turbulence is low and air is, for example, driven by temperature differences that generate free convection, particles display the lowest deposition rates. The deposition rates for all diameters increase with higher air velocities, which can be produced by wind or mechanical ventilation. Deposition rates are also smaller when the surface to volume ratio of the room is small, i.e. when the room has a small surface in relation to its volume. As a general rule, small volumes such as display cases and boxes will have larger S/V ratios than large galleries, but one should bear in mind that the number of objects (e.g. furnishings and exhibits) present in the room will also increase the S/V ratio, and thus increase deposition. Similarly, the roughness of surfaces favours deposition [76].

The applicability of the Nazaroff deposition model has been extensively proven in experimental investigations of particle deposition in a range of environments [77,78], rough surfaces of different materials [79], rooms with fans, and furnished or unfurnished rooms [80].

Re-suspension

Once deposited, PM is adhered to surfaces by adhesion forces that can be orders of magnitude higher than gravity [71], and of which Van der Waals adhesion is the most relevant [81]. Changes in air flow conditions can eventually compensate these adhesion forces and re-suspend the deposited particles. Re-suspension rates are strongly dependant on particle diameter. Larger particles are re-suspended more easily. In some museum environments, particles of $>1 \mu\text{m}$ appear only during museum opening hours due to re-suspension caused by visitors. These particles redeposit gravitationally as soon as the museum is closed [17]. This type of behaviour has been studied for a long time, and common indoor activities such as walking and vacuum cleaning have been associated with re-suspension of particles $>1 \mu\text{m}$ [82], and have been found to increase particle concentrations up to 7 times the background concentration [83]. Re-suspension due to inappropriate cleaning habits has been found to account for the spatial distribution of particles in a monastery which displayed an otherwise very stable indoor environment [84].

This mechanism is very dependent on particle size, and $<1 \mu\text{m}$ particles are rarely affected. Furthermore, re-suspension affects only those particles that are deposited on the floor or the objects involved in the human activity that causes it, such as furniture. The fraction of fine particles involved in the deposition and re-suspension cycle could be expected to be negligible, although there appears to be no relevant experimental research about re-suspension of the accumulation size mode. Nonetheless, re-suspension is a phenomenon that has been extensively modelled [85-87], and it is possible to assess the re-suspension rates of fine particles mathematically.

Figure 9 shows the solution of one of these models, the empirical correlation recommended by [88], along with deposition rates calculated with the Nazaroff equation. A friction velocity of 1 m/s has been used, a relatively high air velocity for an indoor environment which may represent an extreme situation, e.g. the opening of a window with a strong air current. Note that particle re-suspension decays exponentially with time since the moment in which the re-suspending event begins. Consequently, even though re-suspension might exceed deposition when air starts moving, the re-suspension rate rapidly decreases below the deposition rate. The overall particle flux is positive towards surfaces. It can be concluded that as an approximate guideline, re-suspension rates for

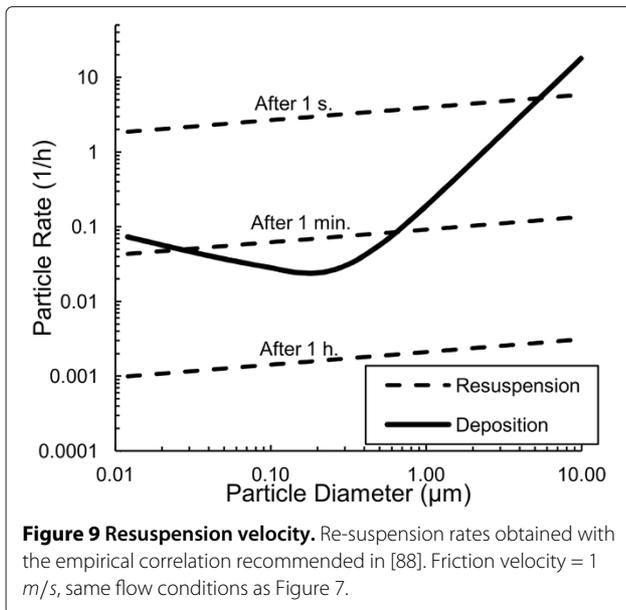


Figure 9 Resuspension velocity. Re-suspension rates obtained with the empirical correlation recommended in [88]. Friction velocity = 1 m/s, same flow conditions as Figure 7.

submicron particles indoors will be orders of magnitude smaller than deposition rates, unless air speed is subject to frequent fluctuations due to wind or mechanical ventilation.

Coagulation

Some authors attribute the measured particle decay in experimental conditions exclusively to deposition while others do to coagulation as well [89]. Naturally, the fraction of particles in a given size fraction that collide or deposit depends largely on the number of particles and on the proportions of the room, specifically on the surface/volume ratio. In some cases either the former or the latter process may be negligible. But in broader terms, it is clear that the temporal evolution of particle number concentration cannot be fully understood without taking both processes into account. In a small chamber with a high concentration of diesel particles (from 8.11×10^5 to 84.3×10^5 particles/cm³, which compares to the range of 1×10^5 to 7×10^5 , concentrations which can be found in street canyons in Stockholm [90]), with diameters from 15 to 670 nm, particle size increased up to a factor of 2.6 during the experiment due to coagulation [91]. An increase of particle size up to 60% has been also found in the case of tobacco smoke during the first 30 min after smoking a cigarette [92]. However, it is rare to find such high concentrations in indoor environments. It has been experimentally demonstrated that coagulation can be neglected in comparison to deposition when total suspended particles (TSP) concentration is under 1×10^3 particles/cm³, and for ultrafine particles ($<0.1 \mu\text{m}$) it is only relevant above 1×10^4 particles/cm³. While particle deposition

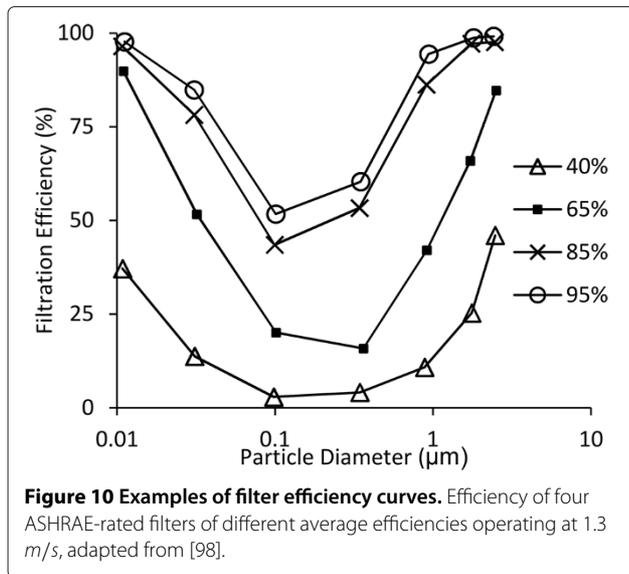
occurs at a constant rate independent of the particle number, coagulation is a second order process that depends on the square root of particle number concentration. This is described by the following equation [93]:

$$\frac{\delta n}{\delta t} = -Kn \quad (1)$$

where n is the particle number and K the coagulation constant. The difference in behaviour (linear for deposition and quadratic for coagulation) allows us to appreciate the effect of both processes on particle decay. Other authors have acknowledged the importance of coagulation as a relevant removal process. It has been found to account for up to 80% of particle loss in a small chamber (1.6 m^3) with steady air, with deposition removing only from 10 to 15% of the paper ash particles used (average particle diameter of $0.069 \mu\text{m}$). This situation changes under stirring, in which case deposition may account for 50% of the removal in the beginning of the experiment and up to 90% at the end [89], as coagulation rate gradually reduces as particle number decays. These results are in agreement with [94], where it was estimated that coagulation could remove from 40% to 70% of the environmental particles in a street canyon with a low wind speed (2 m/s) and around 20% at higher wind speeds (8 m/s), and with the experimental results of [95], who found high coagulation rates in rooms with low air exchange rates. All the mentioned studies focus on particles smaller than $1 \mu\text{m}$, since the smallest particles are more likely to coagulate, not only because of their higher mobility, but because they are typically present in higher number concentrations [73]. Coagulation is known to be fairly independent of particle composition and air relative humidity [96]. There is little doubt that coagulation is a relevant removal process in enclosed or semi-enclosed and highly polluted environments [97], but it is also true that it may be negligible in most heritage environments.

Filtered removal

PM can be removed using a filtration system. There are a number of filter configurations. Air inlet and outlet can be both filtered, or air can be filtered and recirculated into the room in a closed system. The efficiency of such filters is well known and its calculation as a function of particle size is standardised throughout the industry. Commonly, efficiency is expressed using the Minimum Efficiency Reporting Value (MERV) ranging from 1 - 20, 1 - 4 being common values for domestic filters capable of retaining only the largest particles ($>10 \mu\text{m}$) and 17 - 20 are typical values for fine filters used in pharmaceutical or electronic applications, capable of retaining submicron sized particles ($<0.3 \mu\text{m}$), Figure 10 shows the efficiency of some American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE) rated



filters [98]. The ASHRAE recommends museums using filters with a MERV from 7 - 11 [99]. Particle filtration is governed by the same physical mechanisms as infiltration through cracks, thus the expected removal efficiency for each particle size will be inversely proportional to the particle penetration factor. As discussed, fine particles deposit through Brownian diffusion and coarse particles are easily intercepted by impaction, while the accumulation mode exhibits the lowest removal efficiency. It is also worth remarking that an increase of air flow rate will improve the removal of fine particles, but would be less effective as particle size increases, as larger particles are less affected by airflow. The accumulation mode of DPM are the particles, which are removed less efficiently by ventilation systems.

The consequences of deposition of fine particles

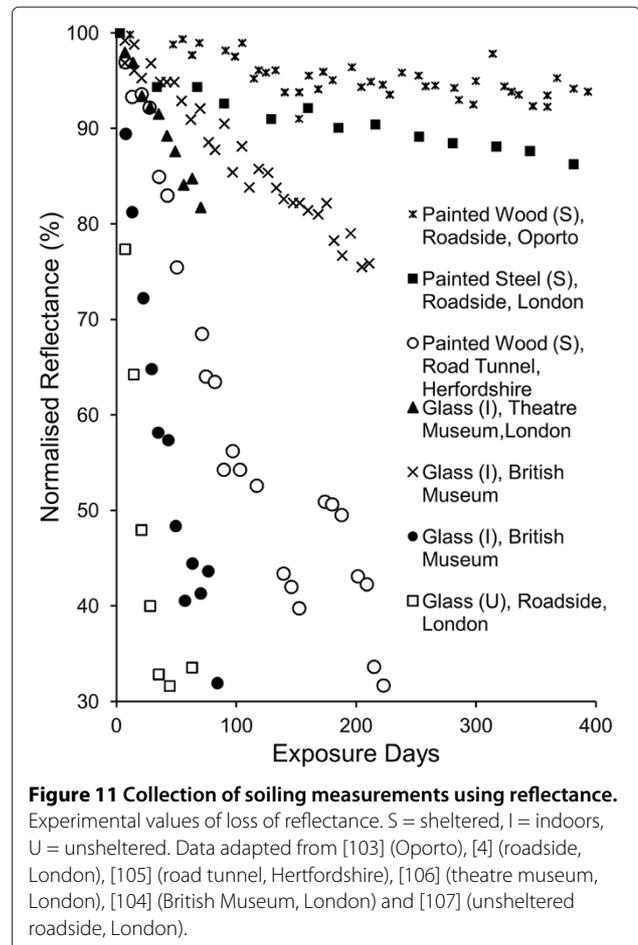
Deposition is a three-fold problem. First, mere deposition (or “dry” deposition) can cause area coverage and have a visual effect on the soiled object, a “visual nuisance”, as it has been qualified in some of the most relevant investigations [100,101]. Secondly, frequent or intense cleaning might have a negative effect on the underlying surface, as well as being a cost-intensive process. And finally, the deposited particles might interact chemically with the surface, creating a damage layer and producing irreversible degradation. Although evidence exists for all these phenomena, research has prioritized soiling on outdoor surfaces and particularly layers produced on stone. The following section attempts to describe these three processes indoors -visual nuisance, chemical damage and damage by cleaning- based on the understanding that evidence is scarce, and in some cases conclusions must be extracted from evidence obtained in outdoor experiments.

Soiling indoors

Figure 11 may convey a sense of the time-scale of soiling processes. It shows experimental values adapted from different publications of the percentage of reflectance lost during environmental exposure. Due to the lack of experimental data on soiling indoors, some sheltered cases are included. The time needed to achieve a 10% loss of reflectance, which corresponds approximately to a change visible by the naked eye [102], is highly variable and ranges from a month to a year. Note also that all soiling processes occur at a varying rate, which tends to be higher during the first weeks of exposure [4,103-107]. This non-linear behaviour is what equations (2), (3) and (4) attempt to reflect.

Soiling is generally assumed to be directly proportional to the loss of reflectance and lightness (or increase in haze) of a material. Several models for soiling are currently under discussion. Historically, the first dose-response functions were developed under the assumption that soiling is proportional to the square root of the concentration of PM.

$$\frac{\Omega}{\Omega_0} = k\sqrt{C_{PM}t} \quad (2)$$



where Ω is an optical property and Ω_0 its initial value, k is a “soiling constant”, t is the elapsed time and C_{PM} is the concentration of PM in the surrounding air. This model was first proposed by Beloin and Haynie in 1975 [108] and has been significantly endorsed by several researchers [10, 103,109]. The second model, also widely used, follows an exponential relation in the form:

$$\frac{\Omega}{\Omega_0} = \exp(kC_{PM}t) \quad (3)$$

which was firstly suggested by Mansfield and Hamilton in 1989 [110] and has also been extensively used [4,111,112]. For some comparisons and critical assessment of these models see [103,104]. Lastly, the most recent research suggested the use of the Hill equation for the development of dose-response functions:

$$\Omega_t = B + \frac{K}{1 + (M/t)^H} \quad (4)$$

where B , K , M and H are constants that define the varying response of soiling with time (see [113] for a detailed explanation). This model has been fitted to experimental data by Lombardo and Ionescu at different occasions [114,115] and has been tested with data collected during the MULTI-ASSESS project [116] in different European locations. A second discussion relates to which concentration should be used as C_{PM} . Some authors have suggested total suspended particles, while others have used $PM_{2.5}$, PM_{10} , DPM or particulate elemental carbon (PEC), given that most of the soiling is due to traffic-generated particulates [110].

Cases of soiling indoors

Soiling outdoors is popularly associated with black stains on faades, while soiling indoors is mostly associated with the deposition of household dust, i.e. coarse particles. In effect, experimental and observational studies are markedly biased towards outdoor blackening and indoor dust, perhaps due to the experimental difficulties of discerning different particle sizes of indoor deposits, especially the smallest. However, it is not difficult to find examples of well visible deposition of fine combustion particles. Some notable examples are found at induction and ventilation outlets [117] or on the murals of a palace in Padova, where inconveniently placed radiators were causing heavy soiling by dust and soot [118]. The darkening in the centre of the murals in Michelozzo’s Courtyard, Florence, Italy, has been attributed to deposition of traffic-generated particles, since measurements of $PM_{2.5}$ display a high proportion of organic and elemental carbon [18]. In some occasions, the damage layers related to the deposition of combustion particles are related to the past use of the building, and not to modern traffic emissions e.g. in the Buddhist statues of the Yunguang Grottoes, China [119].

“Ghosting”: a particular deposition event

A specific indoor discoloration event related to heating points and the presence of semi volatile organic compounds (SVOCs) has been repeatedly reported in the literature under various names, which include “ghosting” [120], “black magic dust” [121] and “fogging” [122]. Even though there have been no specific mentions of the phenomenon in a heritage context, it is clearly not exclusively found in dwellings “ghosting” deposits differ from dry deposition by the presence of droplet-like particles, and a layer of condensed SVOCs [123]. The phenomenon is triggered only under certain conditions: emission of SVOCs (e.g. from refurbished materials), low ventilation rates, high temperature gradients (e.g. above lamps or radiators), and, naturally, the presence of PM. The dark appearance of stains is caused by the presence of elemental and organic carbon agglomerates, but coarse dust particles rich in Ca and Fe can be present as well [123]. Even though efforts have been made to isolate the causes that can initiate this soiling event, all attempts to reproduce it in experimental conditions have been unsuccessful [124].

Visual consequences

The small size of fine particles has two implications that should be stressed in any discussion of their visibility: their small covering area and their light scattering properties. In fact, an important fraction of fine PM is smaller than the wavelength of light visible to humans (~390 - 750 nm). This, however, does not mean that particles cannot be seen when they accumulate on a surface. Firstly, if enough particles are deposited, the deposit will become visible even if a single particle cannot be seen. For example, candle soot deposits are common, even if the size of particles emitted from a burning candle ranges from 10 - 100 nm [125]. Secondly, fine particles do not deposit alone, and all analyses of deposits have found a certain size distribution. Beyond that remark, it can be added that particles with diameters below the visible range can still scatter light when in suspension or when deposited on transparent materials through scattering in the Rayleigh regime. Several investigations report refractive indices for particles or soot small in comparison with the wavelength of light [126,127]. These aspects; however, have not been researched in the context of heritage science.

While perceivable visually, the effects of soiling are quite difficult to quantify. Recently, attempts have been made to identify thresholds between acceptable and unacceptable levels of soiling on building faades. However, the relations between perception and soiling are complex, as the reaction of the public is not simply proportional to the amount of matter deposited. Soiling can be perceived in some cases as patina, and to a certain degree, it can enhance the appearance of a building [128]. The perceived degree of soiling is also influenced by the cleanliness of the

surrounding environment. Despite these issues, Brimblecombe et al. [100] used on-site questionnaires to identify soiling levels that are publicly unacceptable. Their results show that the public perception measured in terms of perceived lightness is fairly consistent with the perceived need for cleaning and could be used to define threshold doses in terms of environmental particle concentration. Another study [129] shows that soiling is perceived as negative when it interferes with architectural shapes. However, these studies are focused on the darkening of building features outdoors, and it can be argued that they have a limited applicability to indoor deposition.

Some researchers have left aside the complexities of aesthetic judgement, and have concentrated on the limits on the perception of soiling. Bellan et al. [60] have measured the human eye ability to detect soot deposition on flat, plain colour surfaces using printed dots (60 - 160 μm) on white and coloured matte surfaces. Their results show that some observers are able to discern a soiled surface from a clean one when the covered area is just a bit higher than 1% of the total, and that deposition becomes obvious to all at around 9% coverage. The perceptive ability is improved if the soiled surface is observed alongside a clean one, in which case all observers identify soiling when just 3.5% of the area is covered. Experiments with larger dots (0.5 - 1 mm) have led to a threshold of 0.2% area coverage [102]. These results have been of great use for the establishment of guidelines and recommendations, e.g. by [130], since they provide a threshold value in terms of area coverage. But it must be noted that the diameters used in the experiments correspond with the coarser dust rather than with "soot" or fine particles, for which no direct account of their thresholds for visibility has been published.

Recently, Druzik and Cass claimed that some specific paintings were under special risk of soiling [131]. Particularly paintings with large colour fields, like those by Mark Rothko or Franz Kline, were assessed as being more vulnerable to the aesthetic damage due to soiling.

Degradation of soiled objects

It has long been established that particulate pollution from road traffic contributes greatly to the degradation of stone outdoors. The presence of DPM has been related to the decay of carbonate [132] and silicate stones [133]. Also outdoors, several corrosion products of copper were identified on statues where soot was also present [134], but no formal relation was established. Although the effects on materials other than stone are less investigated, it can be expected that the reactive components of DPM will also interact with materials, which are typically displayed and used indoors, such as paper, paint and varnishes, or textiles.

There is an important lack of literature about the effects of particulate deposition on the surface of paper, leather,

textiles, paintings, varnishes and other materials typically found indoors, perhaps due to the complexity of the problem and the great variety of materials involved. A brief list of potential degradation pathways related to particle deposition is available in the literature [17]: (i) S-rich material (such as DPM, which contains oxidised sulfur compounds [41]) can cause discolouration of paintings; (ii) ammonium sulfate can induce bloom on varnish. Ammonium sulfate is a "secondary aerosol" (i.e., formed in the atmosphere), but it often coexists (and even aggregates) with carbonaceous particles [135]; (iii) The presence of CaSO_4 favours the adsorption of soot; (iv) Fe-Rich particles can catalyse the oxidation of SO_2 to H_2SO_4 . Aged diesel particulate matter is hygroscopic [136], and therefore can favour the adsorption of water that accelerates hydrolytic and oxidative processes, leading to fading of pigments, and degradation of paper and textiles [11]. Despite the lack of systematic investigations, the effects of fine PM deposition have been repeatedly noted by conservators. Damage layers related to black carbon deposition have been detected on indoor murals and wall paintings [137], and on polychromy [138]. The word "black crust" is sometimes used to describe these damage layers found indoors, but it is a macroscopic assessment that gives very little information about the origin of degradation.

Conclusions may be extracted from studies carried out with particles of similar composition. The corrosion of zinc and steel has been studied in relation to the presence of deposited particles derived from the combustion of oil (which may be comparable to diesel fuel) and coal [139]. The authors concluded that in relatively unpolluted atmospheres inert particulates can induce corrosion in zinc and mild steel due to differential aeration, an effect which is masked when the overall corrosion rates increase. Oil-ash particles were also found to be much more corrosive than coal-ash particles. Although far from the heritage field in terms of temperature and concentration, some investigations have demonstrated that DPM leads to severe degradation of ceramic filters used in engine exhaust tubes [140].

Cleaning

Cleaning of soiled surfaces can induce undesired degradation. It is well known that cementation of coarse dust increases the difficulty of removal [141]. DPM behaves in a similar way, due to its ability to penetrate into pores and its potential chemical interaction with the surface. The National Trust's Manual of Housekeeping states that dry cleaning methods, such as brushing, vacuum cleaning, or even the use of erasers, might be insufficient, and that the deposition of soot can produce "disfiguring, virtually indelible staining" [142]. In cases of extreme soiling, it has been reported that vacuuming removes only the loose deposits of smoke particulates, and that wiping may

further attach particles to porous surfaces [143]. These difficulties have prompted the use of laser cleaning methods; however, laser removal of particles from organic materials have been found to result in yellowing of the underlying surface [144,145].

Conclusions

There is a certain ambiguity of the terms used to describe indoor PM in the heritage literature. There has been some discussion on how the staining of façades should be described (black crust formation, staining, darkening, blackening, soiling) [100,146] but this discussion has not taken place for indoor heritage. As a consequence it is not clear what the word “soot” refers to in some cases. It can refer to carbon-based agglomerates in suspension, or the same particles deposited on a surface, or a black stain of unknown composition but of “carbonaceous” appearance, which might also be only a surface deposit. There is a need for the development of a more accurate terminology that makes a clear distinction between suspended fine particles (DPM, combustion-derived or even soot), dry deposition of these particles without further effects and removable with cleaning (which could be called darkening, or soiling), and the degradation layer formed due to the interaction of the deposit with the underlying surface.

PM monitoring in heritage sites is generally focused on coarse dust, and the two most frequently measured particle types are PM_{10} and $PM_{2.5}$, which include particles up to 2.5 and 10 μm . This standard, however, has limitations. Measurements of $PM_{2.5}$ sum up some particles from the coarse mode ($>1 \mu m$) and some from the fine mode ($<1 \mu m$) and therefore these values do not help to identify the fractions of fine and coarse particles, which would enable appropriate action to be taken. Complete size-resolved measurements of particle concentration would provide more information on the likely source and typology of particles; however, measurement of size distributions requires costly equipment. A much more informative and cost-effective measurement would be PM_{10} and PM_1 , or PM_{10-1} and PM_1 . In a heritage site situated in an urban environment, for example, these values would provide a useful estimate of particles as a consequence of traffic emissions that penetrate into the building.

The formation of black stains in the presence of SVOCs (“ghosting”), or the emission of fine and ultrafine particles when indoor dust is in contact with warm surfaces are phenomena that have been repeatedly observed in indoor environments. It is unknown what the impact of these soiling events is, and whether in some cases they are wrongly attributed to outdoor sources.

Much is known about the aerodynamics of fine PM. The accumulation mode (0.1 - 1 μm), due to its size, displays low deposition rates, low re-suspension rates, and

a high penetration efficiency through cracks and filters. Low deposition rates have different implications. Deposition will be a slow process, but it will occur eventually if particles are not removed. They will distribute evenly around the space, depositing far from the source, and will reach areas in walls and ceiling that are difficult to access. Low re-suspension rates, in combination with a small size that favours penetration into porous surfaces, will lead to difficulties with cleaning and irreversible soiling.

Less is known about what occurs after soiling. There is a significant disproportion between the detailed knowledge of the aerodynamics of fine particles, and what is currently known about the chemical effects of the most common particle types and the potential degradation of soiled (heritage) surfaces. The scarce evidence available is just enough to assess that DPM and other particles derived from combustion can have an active role in the degradation of materials beyond soiling. Considering the costs associated with cleaning, it is important to know if removal of deposited fine particles should be a priority. Risk assessment cannot be based solely on the spatial distribution and deposition rates of fine particles. There is a need for research into chemical interactions between the most common fine particulates (DPM and other combustion-derived particles) and different materials that represent indoor heritage surfaces.

Abbreviations

DPM: Diesel Particulate Matter; EC: Elemental Carbon; I/O: Indoor/Outdoor Ratio; MERV: Minimum Efficiency Reporting Value; OC: Organic Carbon; PEC: Particulate Elemental Carbon; PM: Particulate Matter; SVOCs: Semi-Volatile Organic Compounds; TSP: Total Suspended Particulates; UFP: Ultra-Fine Particulate matter.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

JGB critically analysed the literature and drafted the manuscript, MS co-drafted the manuscript. Both authors co-conceived of the study, participated in its design, read and approved the final manuscript.

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