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Ten Questions Concerning Indoor Ultrafine Particles

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Abstract
Ultrafine particles (≤ 100 nm in diameter) are the smallest condensed-phase materials in air. By number, they are the most abundant particles both outdoors and indoors. Because of their small size, they contribute little to mass concentrations of fine particles, a regulated air pollutant with large associated health risks. Inhaling ultrafine particles raises health concerns, in part owing to their ability to migrate from the site of deposition in the respiratory tract. Buildings provide partial protection against ultrafine particles of outdoor origin. The degree of that protection is broadly variable, influenced by particle size and by building design and operational factors, such as ventilation rates and the efficiency of particle filters. Because of the large proportion of time spent by people indoors, much exposure to ultrafine particles occurs in buildings, even for particles of outdoor origin. Diverse indoor sources, including cooking and the use of candles, contribute materially to indoor ultrafine particle concentrations. Indoor sourced ultrafine particle concentrations are also influenced by building design and operational factors. Whether of indoor or outdoor origin, dynamic processes influence the concentrations, sizes, and chemical composition of ultrafine particles. Such processes include deposition to indoor surfaces, coagulation, and sorptive partitioning of semivolatile organic compounds. Advancing scientific and technical knowledge about building factors and dynamic processes that influence indoor ultrafine particle concentrations can contribute (a) to better understanding of associated health risks and (b) to the more effective design and implementation of mitigation strategies and technologies.

Keywords: Air pollution, controls, exposure, health risk, infiltration factor, sources

1. Introduction
Among airborne particles, the smallest size category is termed "ultrafine," ranging in size from a few nm up to 100 nm in diameter. An authoritative review of indoor particulate matter published in 1996 barely mentions ultrafine particles.[1] However, according to Web of Science, in the past three decades, more than a thousand research articles have been published that include both terms "indoor" and "ultrafine" among their topics. This surge in research attention is driven by several factors, including the dominance of indoor environments as sites of exposure, the potential for adverse health effects resulting inhaling ultrafine particles, and the emergence of technology to enable and facilitate measurement.

Using a question-and-answer format, this paper reviews the state of knowledge about ultrafine particles in commonly occupied buildings. To begin, the reasons for concern about ultrafine

particles (UFP) are explored, focusing on the importance of indoor environments as a site of exposure. Measurement techniques are then reviewed. Sections 2.3 and 2.4 explore the health significance of UFP, respectively addressing the fate of inhaled particles and the evidence concerning the health risks of exposure. Then attention shifts to explore features of the built environment as a site of exposure. Sections 2.5-2.9 respectively address these topics: the degree of protection provided by buildings against UFP of outdoor origin, indoor UFP sources, dynamic behavior affecting ultrafine particles indoors, the proportions of overall exposure that occur indoors, and available mitigation technologies to reduce exposures. The final section looks to the future for ultrafine particles as an indoor air quality concern.

2. Ten questions: asked and answered

2.*1. Why are indoor ultrafine particles important?*
Multiple lines of evidence indicate that inhaling ultrafine particles poses health risks. A large proportion of ultrafine particle inhalation occurs indoors.

The health risk evidence is outlined here and addressed more thoroughly in Section 2.4. As a preamble, for typical indoor and outdoor conditions, the ultrafine particle size range dominates the number-weighted size distribution of airborne particles, contributes substantially to the total surface area, and yet represents a small fraction of particle mass. To the extent that chemical toxicity associated with the bulk of particulate matter causes health harm, ultrafine particles are likely to be less important than the larger fine particles. On the other hand, if health concerns are associated with particle surface properties or especially if they are influenced by the number of inhaled particles that deposit in the respiratory tract, then the ultrafine mode might be very important. Kennedy has noted that "oxidative stress and inflammation are leading contenders" for processes by which particle exposure contributes to adverse health effects.[2] Ibald-Mulli et al. have noted that ultrafine particles deposited in the alveolar region may not be so effectively removed by phagocytosis.[3] The slower rate of clearance by this defense mechanism allows for the possibility of amplified "interactions with interstitial and endothelial cells." Also, the small size of ultrafine particles allows for their translocation from the deposition site to other locations in the body. Delfino et al.[4] stated that "It is likely that redox-active components in UFPs from fossil fuel combustion reach target sites in the lungs, vasculature, and heart to induce inflammation and oxidative stress." Oberdörster et al. noted that the central nervous system "can be targeted by airborne solid ultrafine particles" via deposition on the olfactory mucosa of the nasopharyngeal region of the respiratory tract followed by translocation along the olfactory nerve.[5] Because of the relatively high proportion of surface molecules in the smallest ultrafine particles, such particles can exhibit chemical properties that differ from those of bulk materials of the same chemical composition. Exposures indoors to engineered nanoparticle materials also may occur owing to their incorporation into consumer products.[6]

Population studies support the concern that exposures associated with outdoor ultrafine particles pose health risks. In a systematic review, experts "judged the small database of epidemiological studies supplemented with experimental studies sufficient to quantify effects of UFP on all-cause mortality and to a lesser extent hospital admissions." [7] As a specific

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numerical result from that study, among 11 experts, the overall median estimate was 0.30 for the "estimated percentage decrease in all-cause mortality associated with a decrease of 1000 particles/cm^3 [in the concentration of] ultrafine particles." Another expert assessment found, with medium to high confidence, a likelihood of an "independent causal relationship between increased short-term UFP exposure and increased all-cause mortality, hospital admissions … [and] aggravation of asthma." [8] A recent review of epidemiological studies concluded that "evidence suggests adverse short-term associations with inflammatory and cardiovascular changes, which may be at least partly independent of other pollutants. For the other studied health outcomes, the evidence on independent health effects of UFP remains inconclusive or insufficient." [9]

The second major point, that a large proportion of ultrafine particle inhalation occurs indoors, hinges on three lines of evidence. First, most of people's time is spent indoors, and so most inhaled air is indoor air. Second, although buildings provide some protection against particles of outdoor origin, that protection is incomplete. The time-averaged concentration of ultrafine particles in indoor air that originated outdoors can be a substantial fraction of the time-averaged outdoor concentration. Third, various indoor processes and activities can lead to the direct emission or indoor formation of ultrafine particles. The first idea is developed further in the following paragraph, and the latter two ideas are respectively addressed in Sections 2.5 and 2.6.

Data on time-activity patterns, i.e., budgets describing where people spend their time, have been collected for exposure assessments since the late 1980s. The most highly cited work reports a probability-based survey of 9386 subjects in the United States, with sampling conducted during the years 1992-1994.[10] Surveys have also been conducted in California,[11] Germany,[12] Canada,[13,14] South Korea,[15] and seven European cities.[16] Although the details vary, data are largely consistent across the different populations sampled. The average time spent indoors at home varied between 58% (seven European cities[16]) and 70% (Canada[14]). For studies that reported time outdoors, the average proportion was in the range 6-8%.[10,11,13,14] Hence, the proportion of time spent indoors at home averages about an order of magnitude larger than the proportion of time spent outdoors.

2.2 How are ultrafine particles measured?
Ultrafine particles are commonly measured using particle number concentration as a surrogate, with detection occurring either optically in condensation particle counters (CPC)[17] or electrostatically.[18,19] Size-resolved concentrations can be measured, for example using a scanning electrical mobility spectrometer[20] to size-sort particles prior to detection with a CPC. Aggregate indicators other than particle number concentration are also measured, such as lung-deposited surface area [21] or the mass concentration of quasi-ultrafine particles.[22]

It is important to highlight that true particle dimensions are not measured, only equivalent properties, which depend on the detection method. The distinction is especially important for solid particles, which may have complex geometries as in the case of soot.[23,24]

Most ultrafine particle measurement techniques provide real-time results with high time resolution relative to the time scale of processes influencing concentrations of indoor ultrafine particles. That feature permits process-oriented data interpretation that is not possible with time-integrated sampling. Conversely, a major limitation is uneven instrumental response with particle properties, especially across the full ultrafine particle size range. Instruments may have different lower size limits for detecting particles, often in the range 6–20 nm. Because a substantial proportion of UFP may be within this size range, the lower measurement limit can influence measured concentrations. Another important consideration is instrument cost and the associated complexity of use.

Most environmental measurements of airborne ultrafine particles have used a CPC. Two major principles form the common foundation on which all CPCs are based. First, under supersaturated conditions, excess vapor will condense onto pre-existing particles, causing them to grow. Second, following condensational growth, these larger particles scatter sufficient light to be detected optically. One important detail in the functioning of a CPC is that particles must be larger than some minimum size to serve as a growth "nucleus." Without condensational growth, ultrafine particles do not scatter enough light to be detected, so particles smaller than the instrument's minimum size limit will not be measured. Conversely, particles larger than the cut-off size grow to an extent that is only weakly dependent on initial size. Consequently, particle sizing is generally not possible with CPCs used alone. The minimum particle size measured with CPCs used in field studies has varied, mainly in the range 3-20 nm.[25-27] The upper size limit detected by CPCs is usually larger than the 100 nm upper bound for ultrafine particles and this mismatch can contribute to measurement error.

Some instruments developed for UFP measurements rely on electrostatic effects for detection.[18,19] Commercialized examples have a minimum detectable particle size of 10 nm. They are reported to have lesser precision than CPCs. On the other hand, not needing a liquid reagent allows them to be more easily used for personal monitoring in exposure studies.[28,29]

Consider some typical features of indoor airborne particles. Assume an indoor ultrafine particle concentration of 10^4 particles/cm^3 and, for simplicity in this scaling exercise, assume that all particles are spherical with 30 nm diameter and 1 g/cm^3 density. Then, each particle would have a mass of 1.4×10^{-17} g, so the total ultrafine particle mass concentration would be 0.14 µg/m^3, much smaller than typical indoor fine-particle mass concentrations.[30] The surface area per particle would be 2.8×10^{-11} cm^2, so the area concentration associated with ultrafine particles would be 0.28 cm^2/m^3. Adults inhale about 15 m^3 of air per day. If that quantity of air were to be inhaled in these conditions, then 150 billion ultrafine particles per day would be inhaled with an aggregate mass of 2 µg. The inhaled ultrafine particle surface area would be 4 cm^2 per day. Relative to the other particle size modes, under common indoor conditions, ultrafine particles dominate particle count, contribute relatively little to airborne particle mass, and possess a similar level of suspended surface area as do other fine-mode particles.

Figure 1 displays an example of a representative polydisperse distribution for the atmospheric aerosol. The same major points made in the prior paragraph can be inferred from Figure 1 for

these more realistic conditions: ultrafine particles dominate the number distribution and particles larger than the ultrafine mode dominate the volume (and therefore the mass) distribution.
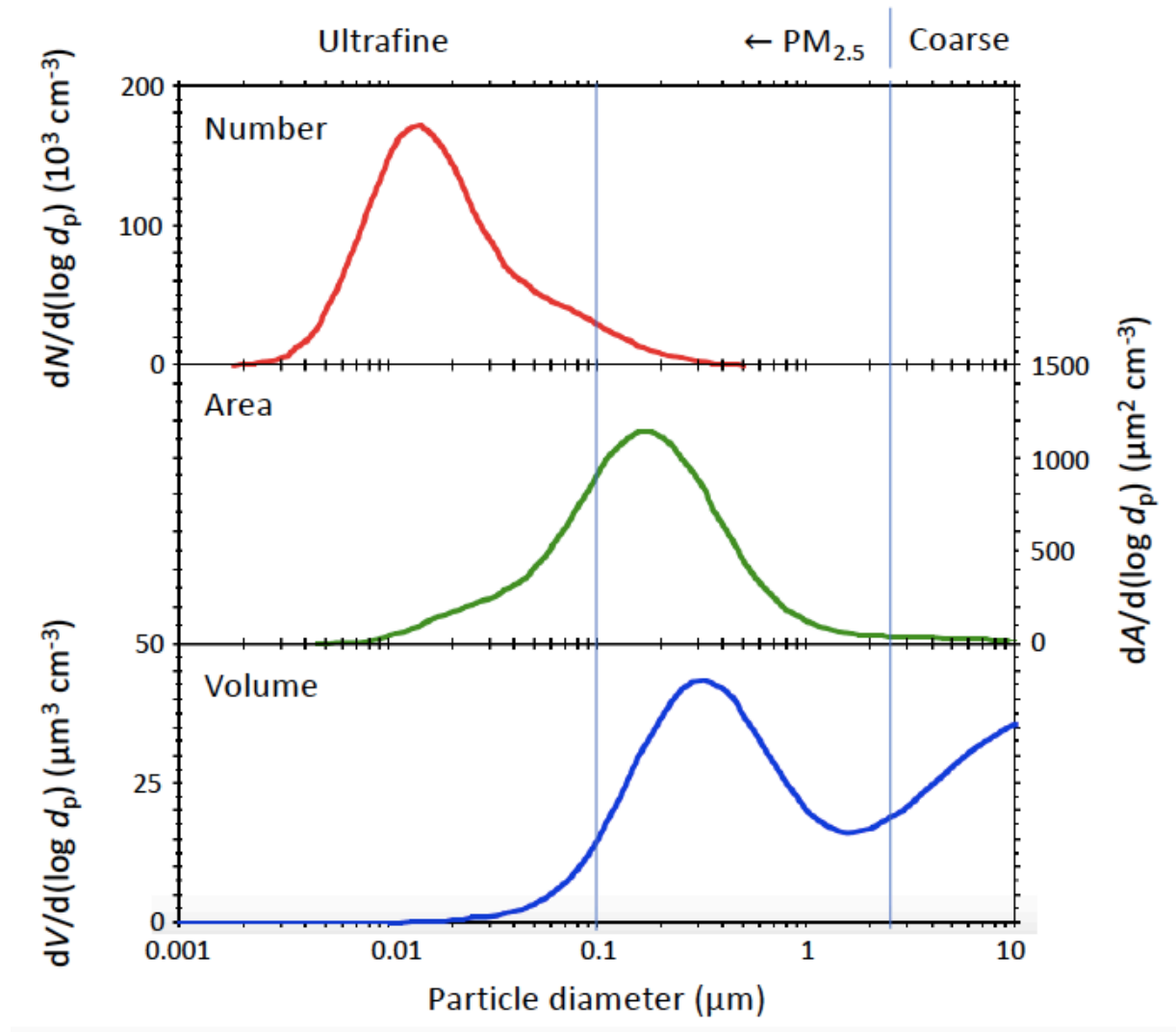

Figure 1. Particle size distribution intended to represent typical conditions for urban outdoor air. The three frames depict the same size distribution weighted respectively by particle number, surface area, and volume. The distribution is constructed as the sum of three lognormal modes, with respective number concentrations of 99300, 1110, and 36400 cm^{-3}; respective modal diameters of 13, 14, and 50 nm; and respective logarithms of the standard deviation of 0.245, 0.666, and 0.337.[31] Considering all particles smaller than 10 μm, the total particle number concentration for this distribution would be 137,000 cm^{-3}, of which 95% are in the ultrafine mode. The total surface area concentration is 1100 μm^2/cm^3, of which 31% (3.4 cm^2/m^3) is associated with ultrafine particles. The total volume concentration is 60 μm^3/cm^3, of which 5% is associated with ultrafine particles. Assuming unit particle density, the PM$_{2.5}$ mass concentration for conditions depicted would be 43 μg/m^3 and the UFP mass concentration would be 3.2 μg/m^3.

2.3. What is the fate of inhaled ultrafine particles?
Inhaled ultrafine particles can deposit throughout the respiratory tract. Subsequent behavior varies with where deposition has occurred, and with the size and chemical properties of the particles. In considering the fate of inhaled ultrafine particles, it is convenient to consider two sequential stages: (a) deposition in the respiratory tract followed by (b) transformation and transport of the deposited particles.

For evaluating deposition, it is useful to divide the respiratory tract into three zones:[32,33]
• Extrathoracic region: from where the air crosses into the body (nose or mouth) to the top of the trachea. It is also known as the NOPL (naso-oro-pharyngeal-laryngeal) region.
• The tracheobronchial (TB) region: branching airways, which increase in number and decrease in size from the trachea to the deep lung.
• The alveolar or pulmonary (P) region: the deepest part of the lung. The primary gas exchange function of the lungs occurs here, in small sacs known as alveoli.
The TB and P regions comprise a branching network of tubes with regular dichotomy. In all, the human lung divides into approximately 23 generations between the trachea and the most distal alveolar-lined airways.

Particle size affects both the overall likelihood of deposition and where that deposition occurs. Brownian diffusion dominates as the transport mechanism that controls UFP deposition. The Brownian motion of an ultrafine particle causes it to deviate from the fluid streamline along which it travels. Any inhaled ultrafine particle that collides with a respiratory tract surface will adhere there.

One often reads that smaller particles are more harmful because they penetrate more deeply into the respiratory tract. For ultrafine particles as a class, such statements are wrong: the smallest UFP deposit most readily in the upper airways. The larger UFP penetrate more deeply. In fact, regional deposition exhibits moderately complex dependence on particle size (Figure 2). A striking feature of each of the two lower frames in Figure 2 is the peak in deposition fraction. In the TB region, the peak occurs at about 4 nm whereas in the pulmonary region it occurs at about 20 nm. It is noteworthy that the peak in the alveolar region corresponds approximately to a peak observed in the number weighted size distribution of atmospheric particulate matter (Figure 1).[34,35]

These peaks in the deposition fraction traces reflect the combined influence of two conditions. First, inhaled air passes through the three regions in succession, such that the "respiratory tract acts as a serial filter system."[36] Second, larger ultrafine particles move more slowly by Brownian motion than smaller particles and are therefore progressively less likely to deposit anywhere in the respiratory tract during inspiration. For conditions depicted in Figure 2, the total deposited fraction decreases monotonically from 98% for 1 nm particles to 30% for 100 nm particles.

After depositing in the respiratory tract, the subsequent fate of ultrafine particles is multifaceted. Influential factors include where deposition has occurred, and what are the sizes

and chemical properties of the deposited particles. Regarding chemical composition, an important consideration is the degree of particle solubility in respiratory tract lining fluids.[37-39] For particles that are poorly soluble, such as elemental carbon and metals, particle integrity may be maintained during biological transport processes.
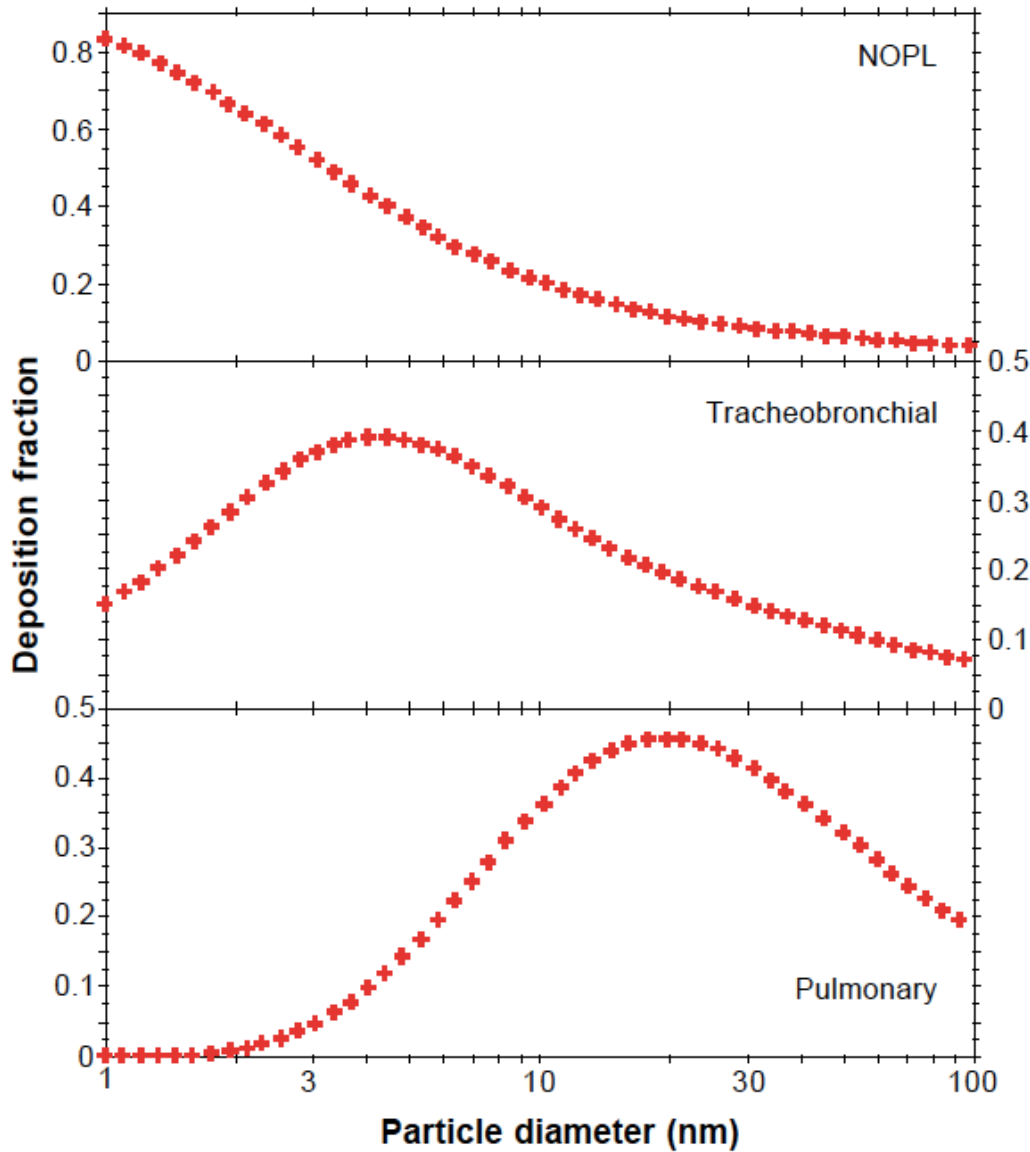

Figure 2. Fractions of inhaled ultrafine particles deposited in the three major regions of the respiratory tract as a function of particle diameter. Conditions are for the case of a healthy adult, seated and breathing through the nose. Results represent the "semi-empirical" modeling efforts of the International Commission on Radiological Protection,[32] as reported by Hofmann.[33]

Clearance mechanisms and persistence time scales vary among the regions of the respiratory tract.[40] Nominally, insoluble particles that deposit in the NOPL region are cleared along with mucus, either mechanically expelled, or swallowed to enter the digestive system. Analogously,

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particles that deposit in the TB region are mainly swept upward by cilia along with mucus and swallowed. The time scales for particle clearance from the NOPL and TB regions are hours to days. Conversely, the pulmonary region is not ciliated. Particles that deposit there can be cleared by phagocytosis with removal time scales potentially extending to months. Additional complications arise when considering the clearance of poorly soluble ultrafine particles. It has been suggested that "the large number of ultrafine particles being inhaled and widely spread over the alveolar epithelium may cause a diffuse, non-localized signal to resident alveolar macrophages which prevents them from directed migration toward ultrafine particles not closely in their neighborhood." [3]

Because of their very small size, insoluble ultrafine particles "appear to translocate readily to extrapulmonary sites and reach other organs by different transport routes and mechanisms." [41] A demonstrated concern is that UFP may enter the central nervous system "from deposits on the olfactory mucosa of the nasopharyngeal region of the respiratory tract and subsequent translocation via the olfactory nerve." [5] Extrapulmonary translocation of ultrafine particles has been demonstrated in laboratory studies of humans exposed under controlled conditions.[42,43] Evidence also indicates that environmental exposures to ultrafine particles can lead to translocation and extrapulmonary accumulation.[44,45] Even if most insoluble particles that deposit on mucus-lined airways are rapidly cleared, the minority that undergo translocation to extrapulmonary locations may contribute to health risks.

In summary, for insoluble ultrafine particles, biological and mechanical processes control the clearance and fate of particles after deposition in the respiratory tract. The potential exists for long persistence times of poorly soluble particles in the body with extensive accumulation in various biological compartments. By contrast, "particle compounds which are either lipid or water soluble will be dissolved and rapidly diluted, bound to proteins, often metabolized in the lining fluid and eventually transferred to the blood and lymphatic circulation, undergoing further metabolization or excretion via kidney and urine." [46] Although soluble ultrafine particles may contain toxic chemicals, "their toxic potential is typically considered low due to the high degree of dilution in body fluids." On the other hand, the high specific surface area of ultrafine particles combined with the high proportion of surface-associated molecules enables biochemical properties that are distinct from those in bulk materials, such as the generation of reactive oxygen species.[41]

A potentially important detail is the potential for multiphase particles. Consider, for example, an insoluble core particle of 3 nm that is encased in soluble material causing the total particle size to be 30 nm. If inhaled in uncoated form, the 3 nm particle would have a high probability of depositing in the NOPL or TB region, with the potential for clearance within a day. However, in the form of the larger coated particle, deposition becomes most probable in the pulmonary region. With long biomechanical clearance times, the materials in the soluble shell could dissolve and dissipate, leaving a much smaller insoluble particle, which could more readily cross the pulmonary membranes. Overall particle size governs regional deposition within the respiratory tract. For multiphase particles, if soluble material dissolves faster than the clearance

time scale, then the subsequent fate of an insoluble core depends on its size and chemical properties.

2.4. What health concerns are associated with exposure to ultrafine particles?
Evidence supports concerns that inhaling ultrafine particles indoors contributes to adverse health risks. Yet, there is also substantial uncertainty in important aspects of this evidence. This section summarizes major lines of supporting evidence and highlights unresolved challenges.

To begin, one can look at the voluminous and largely coherent evidence indicating that environmental exposures to fine particle mass concentrations pose major health concerns.[47] An important development in recent decades is recognizing that inhaling fine particles not only can damage the respiratory tract, but also contributes to adverse cardiopulmonary outcomes.[48] Subclinical adverse indicators linked to fine-particle exposure include systemic inflammation, systemic oxidative stress, autonomic dysregulation, vascular dysfunction, and thrombosis.[49]

Many of the mechanisms of concern for cardiovascular diseases triggered by fine particle exposure may also apply for ultrafine particles. Seaton et al. [50] expressed early concern that ultrafine particle exposure may pose substantial health risks. Ongoing research supports the idea that preclinical indicators of adverse cardiovascular outcomes are triggered by exposure to ultrafine particles.[4,51] In addition to cardiovascular concerns, adverse respiratory health outcomes may be caused by exposure to ultrafine particles. An area of special attention has been asthma. An early study conducted in Erfurt, Germany, reported a stronger adverse effect of UFP number concentration than of fine particle mass concentration.[52] Childhood asthma has been the subject of investigations into the effects of UFP exposure,[53,54] with the possibility raised that even prenatal exposure to UFP may contribute to elevated risks.[55]

Epidemiological investigations contribute important clues about the health risks associated with UFP exposure. The first investigation to show that elevated outdoor UFP concentrations were associated with increased mortality risks was conducted in Erfurt.[56] Several other studies published during the last decade suggest that outdoor UFP levels are a risk factor for adverse cardiopulmonary health outcomes.[57-61] The exposure measure in each of these studies is based on the particle number concentration determined outdoors at the city, census tract, or residence of each subject. None of these studies took account of the differences between indoor and outdoor UFP of outdoor origin. Furthermore, no information about exposure to particles of indoor origin could be gleaned from such epidemiological investigations.

Another approach for eliciting health-relevant responses is through short-term manipulation of exposure conditions combined with measuring biomarkers or other acute indicators of the potential for future manifestation of adverse effects. For example, Bräuner et al.[35] exposed healthy young adults in an environmental chamber either to urban near-roadway particles or to filtered air for 24-h periods. They found that DNA damage was associated with oxidative stress, especially in relation to the number concentration of particles in the 57 nm diameter mode. Bräuner et al.[62] applied air filtration in the apartments of 21 couples in Copenhagen, alternating between two days with high efficiency particle filtration and two days of sham treatment. They

reported that "Indoor $PM_{2.5}$ mass, rather than total numbers … of particles had the most important association with [microvascular function]." A similar crossover intervention study [63] found that recirculating filtration units reduced median $PM_{2.5}$ concentration (from 8 to 4 µg/m^3) and particle number concentration (from 7700 to 5350 particles/cm^3), but "no statistically significant effects of filtration … were observed on microvascular and lung function or the biomarkers of systematic inflammation." In a separate study that relied on observational monitoring without manipulation of exposures over two-day periods for 78 residents of Copenhagen, Karottki et al.[64] reported that particle number concentration "mainly driven by candle burning … in the indoor environment may have a negative effect on lung function and markers of systemic inflammation and diabetes." Vehicle emissions are a major source of ultrafine particles in urban air.[65] Hudda et al.[66] exposed 77 experiments to traffic-related air pollutants (TRAP) for 2-h sessions with filtration applied to achieve three different exposure levels, with particle number concentrations (diameters larger than 7 nm) varying between about 2000 and 30,000 cm^{-3}. They concluded that "reducing indoor concentrations of TRAP was effective in preventing acute increases in [systolic blood pressure]." Studies such as these have several important limitations: (a) they are only well suited for measuring acute responses, not long-term consequences of sustained exposures; (b) the moderate number of subjects limits the statistical power as compared to, for example, the much larger populations common in epidemiological studies; and (c) manipulated exposures are generally not able to isolate a single environmental parameter, such as particle number concentration.

A panel of European experts (epidemiologists, toxicologists, and medical clinicians) participated in an elicitation workshop focused on the health consequences of environmental exposure to ultrafine particles.[7,8] Most experts rated as medium to high "the likelihood of an independent causal relationship between increased short-term UFP exposure and increased all-cause mortality, hospital admissions for cardiovascular and respiratory diseases, aggravation of asthma symptoms and lung function decrements." [8] Conversely, a more recent meta-analysis "did not support UFP effects on respiratory morbidity across all ages." [67] This recent study highlighted that "the limited evidence and the large heterogeneity of previous reports call for future exposure assessment harmonization and expanded research."

2.5. How well do buildings protect against exposure to ultrafine particles of outdoor origin?
Buildings provide partial protection against particles that originate outdoors. The degree of protection varies considerably. Influential properties include particle size [68,69] and the volatility of particle components.[70] Building-related factors include air-change rate, the effectiveness of particle filtration,[71] particle deposition rate,[72] and the extent of particle penetration associated with air infiltration pathways.[73,74]

The *infiltration factor*, F_{inf}, quantifies the protection provided by buildings from particles originating outdoors. Assuming a time-invariant outdoor concentration, with stable building operating conditions, and with no indoor particle emissions, the infiltration factor is the ratio of the steady-state indoor concentration to the outdoor concentration. The infiltration factor varies within the range 0–1, with lower values indicating a higher degree of protection. For time-varying conditions, the infiltration factor is approximately the time-averaged indoor

concentration divided by the time-averaged outdoor concentration, absent indoor sources. Because its primary use is for assessing indoor exposure to particles of outdoor origin, it is important that infiltration factors be determined under conditions that are representative of periods of building occupancy and absent any influence of indoor emission sources. In the past two decades many studies have contributed information about infiltration factors for ultrafine particles in commonly occupied buildings, including residences, schools, offices.

For residences, empirical UFP infiltration factors are available for several European cities[75] and for two cities in Canada: Windsor, Ontario[76] and Edmonton,[77] with sample sizes of 30–50 homes in each study. For the European cities, median infiltration factors were 0.19 for Amsterdam, 0.22 for Birmingham, 0.42 for Helsinki, and 0.42 for Athens.[76] For homes in Windsor, Ontario, Canada, the "medians across all homes were 0.16 (summer 2005), 0.21 (winter 2006) and 0.26 (summer 2006)." [76] In Edmonton, "the seasonal-averaged F_{inf} ranged from 0.08 to 0.47 across homes in winter (median = 0.21 …) and from 0.16 to 0.94 in summer (median = 0.57 …)." [77]

For three schools in Athens, respective values of F_{inf} were 0.32, 0.61, and 0.72.[78] Mullen et al.[79] studied 6 elementary school classrooms in California monitored over 18 school days. Using the ratio of school-day-averaged indoor to outdoor concentration as the best indicator, the average ± standard deviation values of F_{inf} were 0.59 ± 0.13. Reche et al.[80] measured indoor and outdoor particle number concentrations in 39 schools in and near Barcelona. Derived values of F_{inf} exhibited a median (interquartile range) of 0.66 (0.5-0.9).

Matson[81] reported UFP measurements in three offices in Sweden that did not allow smoking. The daily mean indoor to outdoor concentration ratio is used to estimate F_{inf}. For the three offices, the average ± standard deviation F_{inf} values were 0.46 ± 0.07, 0.62 ± 0.07, and 0.71 ± 0.13, respectively. Wang et al.[82] reported on 5 years of continuous monitoring in a 2-story, 2700 m^2 commercial building in Rochester, NY. For UFP, the central tendency value of F_{inf} was 0.42. Overall average indoor to average outdoor UFP concentration ratios for three office buildings in Brisbane, Australia, were 0.40, 0.37, and 0.25.[83]

Field experiments have demonstrated a size-dependence of the infiltration factor.[84-86] As expected, the generally observed tendency is for the infiltration factor to decrease with decreasing particle size. However, a minimum I/O ratio at about 20 nm has been reported in two studies.[85,86] Plausible explanations offered for this unexpected observation include low instrument detection limits, unidentified indoor sources, and shifts in particle size attributable to component volatility.[85] Franck et al.[87] observed that the "non-uniform drop in the number concentrations from outdoors to indoors … is accompanied by a shift of the concentration maxima to larger diameters."

An important limitation of the infiltration factor as a concept should be recognized. The original idea of the infiltration factor was based on decomposing indoor particle concentrations into the sum of a contribution from outdoors (infiltration factor times outdoor concentration) plus a contribution from indoor sources.[88] Chemical processes that influence the indoor particle size distribution may not fit well into this scheme. It is feasible, for example, for indoor ultrafine

particle concentrations to be influenced by reactions between ozone penetrating from outdoors and terpenes emitted from indoor sources, such as certain woods or paints.[89] Particulate matter generated in this way is of mixed origin, combining outdoor and indoor precursors. As conventionally defined and applied, the infiltration factor cannot effectively represent the indoor-outdoor ultrafine particle relationship if chemical processes such as terpene ozonolysis are important.

To summarize, the empirical evidence indicates a tendency for higher values of F_{inf} in schools and lower values in residences; higher F_{inf} values occur with windows open as compared to closed-building conditions; and higher values apply for larger particles within the UFP range, whereas F_{inf} values tend to be lower for smaller particles. The higher values in schools than in homes reflects the common condition that schoolrooms are operated with higher air-change rates than homes owing to their higher occupant density. As discussed in Section 2.9, low F_{inf} values can be achieved in schools and in other building types using mechanical ventilation systems with high efficiency particle filtration.

2.6. What are important indoor sources of ultrafine particles?
Sources that emit ultrafine particles into indoor air can be clustered into three overlapping categories: combustion, volatilization/nucleation/condensation (VNC), and oxidation. Examples of combustion include the use of a gas flame for cooking [90] and the use of candles.[91] For VNC, heated surfaces can volatilize organic compounds from surface films that then return, after cooking, to the condensed phase in the form of ultrafine (or fine) particles. Ultrafine particle emissions from heated surfaces have been reported for electric cooktops [92] and during the use of certain electrical appliances.[93] Studies by Wallace and colleagues have illuminated underlying processes.[94,95] Regarding oxidation, the reaction of ozone with terpenes that originate in fragrances or as cleaning-product constituents is another example of an indoor emission source.[96]

The availability of time-resolved indoor concentration measurements has facilitated identifying and quantifying indoor emission sources. Indoor emissions often occur episodically. Their onset is exhibited by a rapid rise in indoor concentrations, often to a level much higher than simultaneously present outdoors. After the emissions terminate, the indoor concentration decays owing mainly to removal by ventilation, deposition, and filtration.

Several studies have assessed the relative contributions to indoor UFP concentrations of indoor sources versus outdoor air.[91,97-100] In residences, it is consistently observed that indoor sources make major contributions to indoor UFP concentrations. In central tendency, time-averaged contributions from indoor emissions are of similar scale as the time-averaged contribution from outdoor air. Episodic emissions from sources such as cooking can produce short-term peaks in the concentration profile during which the indoor source is clearly dominant. In cases with heavy use of indoor sources, such as extensive use of candles, indoor emissions can dominate even on a time-averaged basis. Because many episodic emission events occur because of occupant activities, the contribution of indoor emissions to exposure is amplified above its contribution to the time-averaged indoor concentrations.[101] Indoor sources have also been

identified in other indoor environments, such as schools [96] and offices.[102] However, contributions of indoor emissions in these settings are generally less prominent than in residences.[103]

Tables 1 and 2 summarize much of the literature that characterizes ultrafine particle emissions from common indoor sources, respectively based on field studies (Table 1) or laboratory investigations (Table 2). Each line in these tables represents a different publication reporting independent experimental data.

Table 1. Field investigations of indoor sources of ultrafine particle emissions.[a]

Source	E	S	C	A	Min d_p	Reference
Cooking and related activities						
Cooking (gas range; eggs, chicken, soup)		✓		Ft	17 nm	104
Cooking (gas range; mostly frying)	✓	✓		Fo	10 nm	90
Electric appliances, heated pans				Ft	10 nm	94
Cooking (electric stove)	✓	✓		Fo	15 nm	101
Cooking (gas, boil, sauté, roast, broil)	✓			Ft	6 nm	105
Heating surfaces with sorbed SVOCs	✓	✓		Ft	2 nm	95
Combustion: Candles, cigarettes, incense, mosquito coil, heating						
Tobacco, mosquito coil, joss stick		✓		Ft	17 nm	106
Wood-burning stoves		✓		Ft	6 nm	107
Candles	✓	✓		Ft	2 nm	108
Appliances: Printers, clothes dryers						
Clothes dryer	✓	✓		Fo	10 nm	109
Laser printer				Ft	10 nm	102
3-D printer	✓	✓		Ft	10 nm	110
3-D printer	✓	✓	✓	Ft	1 nm	111
Field monitoring campaigns that quantified observed sources						
Houses: terpenes in cleaning products		✓		Fo	20 nm	112
Houses: cooking, other activities	✓			Fo	7 nm	113
House: cooking, candle, incense, …		✓		Fo	10 nm	114
Houses: cooking, furnace, candles, …	✓			Fo, Ft	6 nm	98
Houses: cooking, clothes dryer, candles, …	✓			Fo	10 nm	115
Schools: cooking, printing, cleaning, …	✓			Fo	6 nm	116
Houses: candles, smoking, cooking, …	✓			Fo	10 nm	99
Houses: cooking, candle, fireplace, …	✓			Fo	10 nm	100

[a] Relevant information reported in the cited reference is indicated by a checkmark: E = quantified emission rates or emission factors, S = size-dependent emissions information, C = chemical properties of emissions. A = experimental approach (Ft = field study that targeted specific source(s) with purposeful experiments, Fo = field study that interpreted observational data). Min d_p = the minimum particle size measured in the study.

Quantifying emissions is a major challenge for indoor source characterization. Ideally, emissions are assessed in such a way as to permit extrapolation to other settings. At a minimum, the total number of ultrafine particles associated with a source event is needed. Information about particle size and chemical composition is valuable. Time-series monitoring data can be interpreted through applying a material balance model to extract quantitative information about emissions.[90] To the extent that the emissions processes are similar, the quantity of emitted UFP from one experiment can be applied to predict concentrations in other settings.

Table 2. Laboratory investigations of indoor sources of ultrafine particle emissions.[a]

Source	E	S	C	A	Min d_p	Reference
Cooking and related activities						
Cooking (gas and electric; various foods)		✓		Lr	10 nm	92
Cooking (gas, electric, toaster oven)	✓	✓		Lr	3 nm	117
Cooking (gas, electric, grilling, frying)	✓	✓		Lr	6 nm	118
Cooking (volatility from grilling, frying)	✓	✓	✓	Lr	6 nm	119
Heated cooking oils	✓			Lb	10 nm	120
Heating surfaces with sorbed SVOCs	✓	✓		Lr	2 nm	95
Cooking (gas, simulated meal for 4)	✓	✓		Lr	10 nm	121
Cooking (gas, electric, simulated meals)	✓	✓		Lr	1 nm	122
Combustion: Candles, cigarettes, incense, mosquito coil						
Candles	✓	✓		Lb	10 nm	123
Incense, candles, mosquito coils	✓	✓		Lb	4 nm	124
Cigarette smoke	✓	✓		Lr	10 nm	125
Candles (stressed burning)	✓	✓	✓	Lr	2.4 nm	126
Candles	✓	✓		Lb, Lr	7 nm	127
Candles (steady burning)	✓	✓	✓	Lb	2.4 nm	128
Candles	✓	✓		Lr	6 nm	129
Oxidation chemistry, such as ozonolysis of terpenes						
Ozonolysis (terpenes, consumer products)				Lr	20 nm	130
Ozonolysis (cleaning agents, fragrances)		✓		Lb	8 nm	131
Ozonolysis (cleaning agents, fragrances)	✓	✓		Lb	8 nm	132
Ozonolysis (limonene)		✓		Lb, Lr	5 nm	133
Ozonolysis (terpenes from wood, paint)				Lb	20 nm	89
Ozonolysis (limonene on surfaces)		✓		Lb	15 nm	134
Ozonolysis (squalene on surfaces)	✓	✓		Lb	10 nm	135
Ozonolysis (mosquito repellents)	✓	✓		Lb	5 nm	136
Oxidation (cigarette smoke)		✓	✓	Lb	14 nm	137
Oxidation (limonene by bleach, light)		✓	✓	Lb	10 nm	138
Floor cleaning products	✓	✓		Lr	4 nm	139
Ozonolysis (skin lipids)	✓	✓		Lr	1 nm	140
Ozonolysis (terpenes from mopping)		✓		Lr	1 nm	141
Appliances: Printers, hair dryers, etc.						
Laser printer	✓	✓		Lb	5 nm	142

Laser printer	✓	✓	✓	Lb	4 nm	143
Appliances (toaster, hair dryer, …)	✓	✓	✓	Lb, Lr	6 nm	93
Laser printer	✓	✓		Lr	4 nm	144
3-D printer	✓	✓	✓	Lb	1 nm	111
Hair dryer	✓		✓	Lb	N/A	145
Laboratory studies of diverse or miscellaneous sources						
Cigarette, air freshener, candle, cooking …	✓			Lr	20 nm	146
Vacuum cleaner motor	✓		✓	Lb, Lr	10 nm	147
Candle, incense, cooking, spray use, …	✓	✓		Lm	5 nm	148
Candle, cooking, hair drying, incense, …	✓	✓		Lr	11 nm	149
Cigarette, incense, vacuuming, cooking	✓	✓		Lm	15 nm	150
Stove use, candle use	✓	✓		Lr	2 nm	151
Heated terpenoids in vaping or dabbing	✓	✓		Lr	5 nm	152

[a] Relevant information reported in the cited reference is indicated by a checkmark: E = quantified emission rates or emission factors, S = size-dependent emissions information, C = chemical properties of emissions. A = experimental approach (Lb = laboratory using a bench-scale apparatus [chamber ≤ 1 m³, or ducted exhaust], Lr = laboratory using a room-scale chamber (≥ 4 m³) or multiroom test facility, Lm = laboratory study with medium sized chamber [1.5-2.5 m³]). Min d_p = the minimum particle size measured in the study.

Even when emissions are quantified, challenges remain in determining the best unit of measure and normalization. Regarding unit of measure, common options are either to specify an emission rate (e.g., number of particles emitted per minute)[115] or to specify an aggregate total number of particles emitted for a given event.[98] Géhin et al.[148] present a strong example of effective reporting of size-dependent particle emission rates from small-chamber laboratory studies. For each activity, they describe the intensity of the activity and report emissions in terms of rates with particle sizing captured as the sum of one to three lognormal distributions. Regarding normalization, for some types of activities or processes, an appropriate normalization can be readily identified. In candle use, one could assess the quantity of UFP emitted per mass of wax consumed.[124] For cooking, in which many factors may influence the emissions rate, it is challenging to report experimental data in a manner that permits application to other circumstances.

2.7. How do dynamic processes influence the size distribution and chemical composition of ultrafine particles indoors?
Three types of processes are relevant. 1. *Deposition* is a net loss process that entails the transfer of an airborne particle to a surface so that it can no longer be inhaled. 2. A particle colliding with and adhering to another airborne particle is termed *coagulation*. By this process the number concentration of particles is reduced, and mean particle size is shifted upward. 3. *Gas-particle phase change* processes may occur, with subcategories including condensation, evaporation, sorptive partitioning, and nucleation. In the context of UFP dynamics, condensation and evaporation are complementary processes that apply when a single chemical species undergoes phase change with any net movement causing an increase or decrease in

particle size. Sorptive partitioning describes a minor species being absorbed into a liquid condensed phase, or a minor species adsorbing to solid particle surfaces. Nucleation is the formation of new particles in air, generally a precipitous process.

Deposition has been represented as a first-order loss process applied to the total particle number concentration. Caution is advised, since the deposition rate varies with particle size, and so the aggregate behavior is sensitive to the particle size distribution. While recognizing these limitations, it is informative that field studies assessing the deposition loss rate for total particle number concentration have yielded results in the vicinity of 1 per h.[76,98,153]

A few field experiments have determined size resolved UFP deposition loss rates. Long et al.[84] inferred particle loss rates for ultrafine diameters in the range 20-100 nm based on monitoring in 9 houses. They reported a relatively weak particle-size dependence for the deposition loss rate, which spanned 0.2-0.4 h^{-1} using nightly average data from all nine study homes. Wallace et al.[154] reported loss-rate coefficients for 13 ultrafine particle sizes spanning 10-100 nm as determined in an occupied townhouse under four experimental conditions. For the "no fan" condition, the deposition loss rate coefficient varied monotonically with ultrafine particle size from 4.1 h^{-1} for 13 nm particles to 0.80 h^{-1} for 91 nm particles.

Regarding coagulation, one should note that there is vastly more surface area on fixed indoor surfaces than associated with airborne particles. A typical value of the surface-to-volume ratio for an indoor space is 3 m^2/m^3; [155] particle surface area concentrations are orders of magnitude smaller. Compensating somewhat for the smaller abundance of surface, the distance that must be travelled in room air to reach a suspended particle is small. Coagulation can be consequential for the smallest ultrafine particles, especially when the fine particle mass concentration is high. To a good approximation, the principal outcome is to transfer nanometer-scaled particles to the sizes where most of the particle surface area resides, generally in the vicinity of 100 nm diameter. Coagulation can affect the fate of nanometer scaled particles, including the likelihood and the location of depositing in the respiratory tract if inhaled and, potentially, the fate in the body after deposition. Once attached, the mobility of the larger particle governs the behavior of the coagulated pair.

Among phase-change processes that apply to indoor ultrafine particles, two categories appear most germane: (1) oxidation-induced nucleation and growth, and (2) a shift in the gas-particle partitioning of semivolatile species caused by changes in environmental conditions.

Terpenes are unsaturated hydrocarbons that react rapidly with ozone. The products of such reactions include molecules with lower vapor pressure and higher polarity than the parent species. When such reactions happen indoors, the thermodynamically favored outcome is for the low vapor-pressure products to condense. One option is condensation on room surfaces, but that is a slow process because of mass-transport limitations. Condensing on pre-existing particle surfaces is more likely because of the proximity of such surfaces. If the rate of product generation exceeds the rate at which condensation onto existing particles can occur, then nucleation may occur, by which new ultrafine particles are formed. New particle formation

creates new surface area available for condensation, shifting the balance between nucleation and growth. A commonly observed outcome is a burst of new particle formation followed by condensation-induced particle growth. The newly formed particles are initially at the finer end of the ultrafine particle size range. Condensational growth occurs, not only across the full range of ultrafine particle sizes, but also on particles larger than 100 nm. From a physicochemical perspective, the condensational growth process blurs the distinction between ultrafine and accumulation mode particles.

Long et al.[112] published the first observational field study that identified nucleation events indoors induced by ozone reacting with terpenes. Since that study, and despite widespread scientific recognition of the potential for ozone to combine with terpenes to form ultrafine particles, only a few observational studies in ordinarily occupied buildings have reported such findings. In one example, Morawska et al.[96] investigated ultrafine particles in classrooms in Australia and observed particle formation events from classroom art activities (painting, gluing, and drawing) as well as from cleaning with terpene-containing solutions. Extensive studies of ultrafine particles in residences provide little evidence for ozone reacting with terpenes to form ultrafine particles under "as lived in" conditions.[91,98-100,114,115]

Nevertheless, at least 20 laboratory studies have been published during the past two decades investigating oxidation reactions as a source of indoor particle nucleation and growth.[130,132-135,137,138,140,156-167] In a typical experiment, the reagents, such as ozone and a terpene source, are introduced into chamber air (either bench scale or room scale) and the particle number concentrations are measured as a function of time, often with size-resolving instruments.

Other studies have demonstrated that certain semivolatile organic compounds can sorb substantially to submicron particles.[168-170] However, because of experimental limitations, the extent to which ultrafine particles are influenced by such processes hasn't been elucidated in field experiments.

2.8. What proportion of ultrafine particle exposure occurs indoors?
Ultrafine particle exposure can be quantified for daily periods with the intensity measured in number concentration units. Specifically, a convenient measure is thousands of particles per cm^3 for hours per day duration. So, for example, if one encounters a time-averaged UFP concentration of 10,000 cm^{-3} over the course of a full day, the exposure would be 240 ($\times$ 1000/cm^3 $\times$ h/d). Presto et al.[171] estimated for the United States a population-weighted mean outdoor UFP concentration of 16×10^3 cm^{-3}. Hypothetical continuous exposure to this concentration would produce a daily exposure of 384 ($\times$ 1000/cm^3 $\times$ h/d).

Total daily exposure can be determined as the sum of the contributions from each microenvironment occupied. A convenient decomposition considers four categories of microenvironments: indoors at home, indoors at another location, outdoors, and in transportation. Then, the total daily exposure is constructed as follows:

$$X = C_h t_h + C_l t_l + C_o t_o + C_t t_t \qquad (1)$$

Here, the terms C and t represent the time averaged UFP concentrations, and the duration of daily time spent, respectively; and the subscripts refer to home (h), other indoor location (l), outdoor (o), and transportation (t) microenvironments. The daily exposure that occurs indoors is represented by the first two terms on the right-hand side of equation (1). The fraction of exposure that occurs indoors, F_{in}, is the ratio of this sum to the total exposure, X:

$$F_{in} = \frac{C_h t_h + C_l t_l}{X} \qquad (2)$$

For the discussion to follow, specific time budgets appropriate to the study under discussion are applied whenever possible. Otherwise, average results reported by Klepeis et al.[10] for the United States will be used: t_h = 16.5 h/d; t_l = 4.4 h/d; t_o = 1.8 h/d; t_t = 1.3 h/d. These data indicate that an average of 87% of time is spent indoors (69% in one's residence), with only 8% outdoors and 5% in a transportation vehicle. Exposure outdoors at the average US concentration of 16×10^3 cm^{-3} for a duration of 1.8 h/d would contribute 29×1000/cm$^3 \times$ h/d to total daily UFP exposure.[10]

Weichenthal et al.[172] measured particle number concentrations during the evening and overnight hours in 36 Canadian homes during the heating season. They found that the average concentrations during the evening hours (1600-2400) and overnight hours (2400-0800), respectively, were 21.6×10^3 cm^{-3} and 6.6×10^3 cm^{-3}. For occupants at home during all these hours (16 h/d), the average contribution of these periods to daily exposures would be 226×1000/cm$^3 \times$ h/d.

Combining residential monitoring with activity logs, Bhangar et al.[98] assessed the residential UFP exposures of 21 occupants of 7 dwellings in Northern California. The arithmetic mean of these household exposures was 296×1000/cm$^3 \times$ h/d, accumulated over an average time at home of 17 h/d. Using a similar approach, Mullen et al.[173] conducted monitoring in four Beijing high-rise apartments with 13 residents. They reported an arithmetic mean for daily integrated residential exposures of 294×1000/cm$^3 \times$ h/d. Bekö et al.[91] conducted a study of household UFP exposure in 56 homes in Copenhagen. The average residential UFP exposure was 667×1000/cm$^3 \times$ h/d, with substantial contributions from candle use. Zhao et al.[100] monitored 40 households in Berlin and Leipzig for a cumulative period of 497 days and reported average household exposures of approximately 200×1000/cm$^3 \times$ h/d.

A few studies have used microenvironmental monitoring to assess particle exposures in schools. Mullen et al.[79] monitored 6 California classrooms, separately reporting concentrations and occupancy durations. The average daily contribution inside classrooms to exposure was 52×1000/cm$^3 \times$ h/d. Rufo et al.[174] sampled 34 classrooms in 10 schools in Portugal. Using a standard 6.5 h/d period of occupancy, the average contribution to exposure in those classrooms was 59×1000/cm$^3 \times$ h/d. Morawska et al.[103] suggested that average classroom UFP concentrations during occupied times are 14×10^3 cm^{-3}. That level, combined with an estimated

average occupancy of 6 h/d for pupils would contribute 84 × 1000/cm^3 × h/d to exposure on school days.

Restaurants can have elevated indoor UFP concentrations associated with cooking emissions. Ott et al. assessed the exposures for 73 episodes of dining at a restaurant.[175] On average, transportation by car, to and from the restaurant, contributed 17 × 1000/cm^3 × h of exposure and the period of dining added another 72 × 1000/cm^3 × h. Note that these values represent exposures per dining event rather than per day.

Using personal monitoring, Buonanno et al.[176] evaluated daily UFP exposures for both partners among 24 nonsmoking couples in central Italy, selected so that all women were homeworkers and all men worked full-time outside the home. The average exposure for women was higher than for men in both seasons, which was attributed to cooking. Winter exposures were higher than summer exposures. The average exposures by season (W, S) and sex (F, M) were as follows, all in units of 1000/cm^3 × h/d: SF — 432, SM — 221; WF — 696, WM — 312.

Bekö et al.[177] conducted a personal exposure study in Denmark that entailed monitoring 59 subjects for 48-h each. For *n* = 50 subjects with sufficiently complete data, the overall average exposure was 373 × 1000/cm^3 × h/d. The average contribution to exposure while at home was 153 × 1000/cm^3 × h/d, and the contribution from other indoor environments was 191 × 1000/cm^3 × h/d. These indoor contributions sum to 92% of the total.

Pañella et al.[178] reported on 1-day of sampling for 100 children (age 9 y) in Catalonia. Average exposure was 298 × 1000/cm^3 × h/d, of which 57% occurred at home, 20% at school, 13% during transportation, and 9% in other settings. Mazaheri et al.[29] reported on an investigation in Heshan, China, of 24 schoolchildren (9-13 y). Their total average exposure was 697 × 1000/cm^3 × h/d, with major contributions from indoors at home (70%) and indoors at school (20%). Zhou et al.[179] reported on a monitoring study involving 22 high school students in Chengdu, China. The average exposure rate was 439 × 1000/cm^3 × h/d. Van Nunen et al.[180] conducted 24-h of monitoring for 132 adults divided among three European cities. The mean reported concentrations correspond to these exposure levels, all in units of 1000/cm^3 × h/d: Basel — 271, Utrecht — 370, and Turin — 482.

These studies substantiate that dominant contributions to UFP exposure occur in residential environments. The primary reasons are two. First, people spend most of their time indoors at home. Second, although residences (and other buildings) provide some protection against outdoor-sourced particles, that protection is incomplete. Furthermore, there are important indoor sources of ultrafine particles, with cooking being especially prominent, although not uniquely so.

A caution in interpreting overall exposure estimates is emphasized: the instruments used in different experiments can vary in the smallest size detected. Most of the UFP exposure studies

have been conducted using instruments with a lower detected particle size of 10 or 20 nm. Information about exposures to ultrafine particles smaller than 10 nm is scant.

2.9. What control measures are available to limit ultrafine particle exposure indoors?
Two broad strategies provide opportunities for limiting indoor UFP exposures: source control and air cleaning. In considering the potential benefits of control measures, it is useful to differentiate between *efficiency* and *effectiveness*. In evaluating an air-filtration system, for example, efficiency would represent the fraction of particles removed from air as it passes through the control device. Effectiveness would represent the extent to which application of the control measure influences an outcome of concern. It could represent the fractional reduction of the concentration in indoor air or the fractional reduction in the number of particles inhaled while indoors. The concept of effectiveness can be applied for source control as well as for air cleaning. These ideas have been discussed in several articles for the specific case of portable air cleaners.[181-184]

2.9.1. Source Control
Indoor UFP source control has been most thoroughly studied for cooking. Among opportunities for improvement are the choice of energy source (electric stoves emit less than gas stoves), the style of cooking (steaming emits less than frying), and the use of control devices for removing emitted particles from indoor air (ducted range hoods can capture and exhaust cooking-generated pollutants).

With respect to energy source, ultrafine particles are emitted from both gas-fired and electric cooking appliances.[92] More than a century ago, it was found that flames generate ultrafine particles.[185] Much more recently we've learned that, even in the absence of combustion, the heating of surfaces can produce ultrafine particles.[95] Studies characterizing UFP emissions from cooking activities have generally reported moderately higher emissions from gas cooking than from electric cooking.[98,115] Particle emissions from cooking with oil tend to increase with increasing temperature.[118,120] The food that is cooked and the style of cooking can influence UFP emissions.[118,186] UFP emission factors are considerably higher for frying (stir-frying, pan-frying, deep-frying) than for processes that used heated water (boiling, steaming).[187]

The performance of range hoods for extracting stove-top emissions including UFP before they disperse in the residence has been extensively studied in laboratory and field investigations.[105,121,188-190] One commonly reported finding is the tendency for emissions from rear burners to be captured more efficiently than those from front burners.

For candles, reducing emissions could be achieved through reduced usage frequency, duration, and/or intensity (number of candles burned). In principle, it should be possible to substitute candles with lower emission rates through improved combinations of wicks and waxes. Perhaps for some, the perceived benefits of candle use could be achieved with modern lighting technologies that can create a desired mood without combustion.

Laser printers and photocopiers have been reported to be potentially important indoor emission sources of ultrafine particles.[144,191,192] The particles originate mainly from nucleation events so that better control of the fusing process, including use of a lower temperature, can decrease emissions.[193] A small proportion of desktop printer units are high emitters.[194] A recent review concluded that the contribution of laser printers "to the overall particles in typical office and home scenarios was low compared with background exposure or other common indoor sources." [195]

2.9.2. Air Cleaning

Common configurations for air cleaning utilize fibrous filters either (a) placed in an air handing system or (b) mounted in a recirculating fan-filter unit. Depending on the airflow configuration, control can apply to particles of outdoor origin alone or (more commonly) to both outdoor and indoor sourced particles. Important performance metrics include the clean-air delivery rate (CADR),[196] the filtration efficiency,[197] and the effectiveness.[181]

Figure 3 illustrates a common situation in which indoor particle control is sought by using a recirculating fan-filter unit. The efficiency of the filter, η, is the fraction of particles removed from the airstream flowing through the control unit at volumetric rate, Q_r. The clean-air delivery rate is the product of efficiency and flow rate through the control device: CADR = $\eta \times Q_r$. Effectiveness represents the fractional reduction in particle concentrations by the addition of the control measure.[183] For the configuration in Figure 3, and for steady-state conditions, the effectiveness, ε, can be derived from a steady-state material balance, yielding this result:

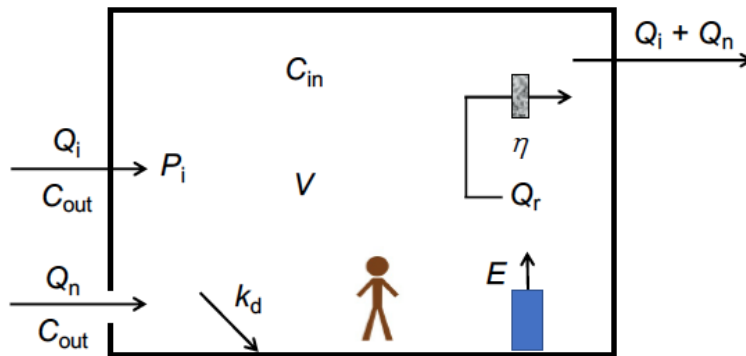$$\varepsilon = \frac{\eta Q_r}{\eta Q_r + (Q_i + Q_n) + k_d V} \qquad (3)$$

Figure 3. Schematic representation of a naturally ventilated indoor environment for assessing the effectiveness of a recirculating fan-filter control unit used to control indoor ultrafine particle exposures. Parameter definitions are as follows. The indoor and outdoor particle concentrations are, respectively, C_{in} and C_{out}. Interior volume is V. Air flow rates are Q_r for recirculating flow, Q_i for infiltration, and Q_n for natural ventilation. Fractional particle removal as air flows through an air-cleaning unit is η. Fractional particle penetration for infiltrating air is

P_i. The first-order loss rate coefficient for particle deposition on interior surfaces is k_d. An indoor source may be present, emitting particles at a rate E.

For a well-mixed indoor environment, equation (3) states that the effectiveness of adding this control measure to remove particles from air is determined by the ratio of the added removal rate to the total rates of removal by all processes. For high effectiveness, the added CADR must be substantial compared to the baseline removal in the absence of control.

Since the pioneering work of Hanley et al.,[197] many studies have reported measurements of the particle-size dependent removal efficiency of commercial filters used in building ventilation systems.[198-204] Across the size range of ultrafine particles, smaller particles tend to be removed with higher efficiency than larger particles. The underlying reason is that diffusion to fibers as air flows through a filter is a key factor influencing particle removal, and the smallest ultrafine particles diffuse much more rapidly than the largest. Other factors studied include the effects on efficiency of airflow rate through the filter, dust loading on the filter, and the manufacturer's rating of filter quality. Li and Siegel [204] measured in situ filter efficiency in residential air handling systems. They noted that "both nominal efficiency and performance change were vastly overshadowed by the wide variety in loading and surface conditions across homes, making it hard to predict filter efficiency in a given home without in situ measurements."

The most efficient filters available for indoor air cleaning are known as "high-efficiency particulate air" or HEPA filters. They have close to 100% removal efficiency across a full range of airborne particle sizes. Lowther et al.[205] reported on chamber experiments assessing the performance of recirculating air cleaners with HEPA filters. As would be expected, in this case the CADR was only weakly dependent on particle size across the measured range 10–500 nm.

Although less used indoors, electrostatic precipitators (ESP) are a viable alternative to fibrous filters for removing particles from airstreams in some situations. However, weak electrical charging of ultrafine particles can pose challenges. For example, one study showed good ESP removal efficiency (> 80%) for particles larger than about 30 nm diameter but decreasing efficiency with decreasing particle size for smaller particles.[206] The ineffective performance for the smallest particles was attributed to a "partial charging regime for particles less than a few tens of nanometers." Another problem for indoor use of electrostatic precipitators is silicon fouling of corona discharge wires originating from volatile methyl siloxanes,[207] a common class of indoor air contaminants.[208] The indoor use of electrostatic precipitators would also raise concerns about their propensity to emit ozone.[209]

During the past decade or so, several field trials have been undertaken to test the performance of particle filtration technologies for reducing ultrafine particles in homes and schools. Residential air-filtration studies have used stand-alone air cleaners with high efficiency filters.[210-213] They have demonstrated moderate effectiveness in reducing indoor particle number concentrations. High levels of effectiveness (> 90%) have been reported in a few studies that have added high-efficiency filters to mechanical ventilation systems in school classrooms.[214,215]

2.10. What is the outlook for ultrafine particles in buildings?

Although all ultrafine particles are miniscule, they span a large range of sizes. The approximately 50× ratio between the diameter of the largest (100 nm) and the smallest corresponds to a vast volume (and mass) difference of > 100,000×. As research continues to generate new knowledge about ultrafine particles, we might anticipate a further division between nanoparticles (e.g., with diameters < 10 nm) and larger ultrafine particles (with diameters in the range 10-100 nm). Nanoparticles have a relatively short airborne lifetime owing to high rates of diffusion to surfaces. A large proportion of their constituent molecules are on or near surfaces, with chemical properties that can differ from bulk condensed-phase materials.[216] When inhaled, the likelihood of deposition is high and is concentrated in the upper airways. Recognizing these and other differences between nanoparticles and the larger ultrafine particles may help efforts to understand sources, dynamic behavior, health consequences of exposure, and mitigation opportunities.

Field studies of indoor ultrafine particles have mainly been conducted using instruments with a minimum particle detection size of 10 nm or 20 nm. There is a dearth of empirical data from observational studies of the smallest ultrafine particles, especially those in the size range 1-5 nm. There is an important research opportunity to better understand indoor concentrations, sources, dynamic behavior, and exposure to the smallest ultrafine particles.

There are sizeable challenges to accurately characterize health-relevant exposure to ultrafine particles. For the built environment, major considerations are the extent to which indoor environments attenuate outdoor concentrations and the role of indoor emission sources influencing exposures. It seems within reach to incorporate knowledge about how building parameters affect the infiltration factor to improve exposure assessments used in epidemiological studies of health effects. The possibility to expand use of personal monitoring as a tool to better characterize exposures in epidemiological studies is tantalizing.

Even without better knowledge about health consequences of exposure, a strong argument can be made in many circumstances to use air cleaning as a precautionary mitigation measure. Health benefits with favorable economics have been demonstrated for the use of better filtration considering only the health risks for fine particle exposure.[217,218] The same approach would reduce exposure to ultrafine particles at no added cost. To ensure that ultrafine particle control is effective, the filter rating systems should be extended into the ultrafine size range.

Some shifts in exposures are likely to result from climate change and the associated societal responses.[219] A greater frequency of wildfires could cause increases in outdoor ultrafine particles, but efforts to protect building occupants against wildfire smoke could compensate. The transition from internal combustion engines to electric vehicles will likely result in lower ultrafine particle concentrations outdoors. Analogously, early evidence of a transition away from natural gas use in homes in favor of electricity should lead to some reductions in exposure. The possibility of much lower UFP emissions from shifts in cooking technologies is indicated by an investigation of indoor air quality in "homes designed to be high-performance

homes in California." [220] That study found that "emissions of ultrafine particles … were about 40 times lower on induction electric cooktops compared with either gas or resistance electric models."

3. Conclusions

Indoor ultrafine particles represent a complex, dynamic system of potential health significance about which much is known and much remains to be learned. This "ten questions" review has explored the state of knowledge about indoor ultrafine particles from the perspective of environmental science and engineering. The ten questions address three major themes — environmental health, pollutant dynamics, and building science — relying on a combination of process-oriented descriptions and empirical evidence. A general summary of the major points follows.

Ultrafine particles dominate number concentrations but contribute relatively little to fine particle mass. Most ultrafine particles are inhaled indoors, in one's residence, because of three factors: (a) most time is spent indoors at home; (b) buildings provide only partial protection against ultrafine particles of outdoor origin; and (c) residences commonly have important sources of indoor ultrafine particle emissions, including cooking.

Inhaled ultrafine particles can deposit throughout the respiratory tract, with smaller particles tending to deposit more in the upper airways and larger ultrafine particles reaching the deep lung. The smaller ultrafine particles, especially if insoluble, have the potential to migrate from the deposition site and become concentrated in extrapulmonary organs. Evidence about health consequences is still in development but is sufficient to justify concern.

The size distribution and chemical composition of indoor ultrafine particles can change rapidly in time and space because of building operations, occupant activities, and environmental conditions. Mitigation through source control and air cleaning can be effective in achieving exposure reductions. Improving knowledge about indoor ultrafine particles will be important as a foundation for decisions and actions that affect exposures and risks.

Biography

William Nazaroff is Distinguished Professor Emeritus in the Department of Civil and Environmental Engineering at the University of California, Berkeley, where he taught and led a research team for three decades (1988-2018). He is a Fellow of the American Association for Aerosol Research (AAAR) and of the International Society of Indoor Air Quality and Climate (ISIAQ). He served as Editor-in-Chief of *Indoor Air* (2009-2018). He is the year 2018 recipient of ISIAQ's Pettenkofer Award for "scientific contributions toward understanding indoor air pollutant dynamics and related occupant exposures." Nazaroff has published extensively on indoor air quality throughout a career that spans more than four decades. Much of his work has focused on indoor particles, including the ultrafine mode.

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