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Developing parameters for multi-mode ambient air models including the nanometer mode

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Abstract. The particle count, surface and mass in an occupied space can be modeled when the HVAC system airflows are known, along with the particle-size distribution for outdoor air, internal generation rates as a function of particle size, and the efficiency as a function of particle size for filters present. Outdoor air particle-size distribution is rarely available, but measures of particle mass concentration, $PM_{2.5}$ and PM_{10}, are often available for building locations. Outdoor air aerosol size distributions are well modeled by sums of two or three log-normal distributions, with essentially all mass in two larger modes. Studies have also shown that some mode parameters are, in general, related by simple functions. This paper shows how these relationships can be combined with known characteristics of $PM_{2.5}$ and PM_{10} samplers to create reasonable inclusive models of outdoor air aerosol-size distributions. This information plus knowledge of indoor particle generation allows calculation of aerosol mass in occupied spaces. Estimation of parameters of aerosol modes with sizes below100 nm and measurement of filter efficiencies in that range are described.

1. Introduction

To calculate the aerosol mass downstream of an air filter, two sets of data are needed. The first is the particulate mass and particle size distribution upstream of the filter. The second is the efficiency of the filter as a function of particle size. The range of sizes for both sets must include all sizes for which there is any significant quantity of particles in the upstream flow. Neither of these two data sets is easy to obtain if the calculation is made for a filter in a real-world building HVAC ventilation system. The outdoor air size distribution changes hour by hour. Even its average pattern will not be known unless some agency samples the air near the system intake, measures the size distribution more or less continuously, and publishes its data.

The particulate concentration and size distribution inside a ventilated space depends on many factors. Among these are the magnitude of the ventilation flows into and out of the space, including recirculation flow; aerosol generation in the space; leaks into and from the space; and the existence of additional filters in flow paths. Some of these factors are easy to quantify, others quite difficult. For ambient aerosols, even the concept of "size" is ambiguous. Only a spherical aerosol particle has a definite, single size. Filtration behavior of cylindrical particles or agglomerates must be expressed in terms of "aerodynamic equivalent diameter" spheres. In addition, filter efficiency also depends on particle density, which can have a wide range of values. For these reasons, standardized filter

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efficiency tests [1]-[4] make use of spherical liquid aerosols and nearly spherical solid aerosols of specified density.

Similarly, studies of the distribution of ambient and indoor particle sizes generally ignore density variations, and accept the sizes defined by whatever principle the distribution-measuring instrument uses. If the distribution is measured by a differential mobility analyzer (DMA) or scanning mobility spectrometer (SMPS), the diameters measured are equivalent to the size measured for a spherical particle with a specified density and a single electron charge. Such diameters are called "mobility diameters". Instruments for optical measurements of size distributions are usually calibrated in terms of the light scattered by spherical polystyrene latex particles; ambient particles are assigned the diameter of a latex particle which scatters the same amount of light. This is then called an "optical particle size". If the size-measurement device is a cascade impactor, the measured mean size on each impactor stage is a function of particle aerodynamic drag and density. Sizes thus measured are expressed as "aerodynamic equivalent diameters", usually for an assumed density of 1000 kg/m^3.

All of these instruments are widely used in studies of indoor air quality (IAQ) and outdoor air quality (OAQ), and to measure the local values of particle mass termed PM_1, $PM_{2.5}$ and PM_{10}. The exact meaning of these three measures of OAQ and IAQ and the observed relations between their values are essential to our discussion. We define them in the next section. References [5] and [6] provide summaries of the operating principles of the various instruments used to monitor ambient aerosol concentration and obtain PM_1, $PM_{2.5}$ and PM_{10}.

Data on filter efficiency as a function of particle size is subject to less ambiguity than ambient particle-size distribution data, provided one is willing to accept that the results of efficiency tests by standard procedures are representative of the results that would be obtained with typical ambient aerosols. Efficiency-vs-particle size data is also likely to be more easily obtained, since all internationally recognized standards require such data as part of the qualification of a filter.

However, obtaining this efficiency-vs-size data may require some persuasion of the filter manufacturer, for filter advertising material is almost always limited to filter "ratings". Ratings are single numbers which place a filter in a category, based on tests performed following the standard. Under EN 779:2012 [1] the rating is based on a single particle diameter, 0.4 μm. ASHRAE Standard 52.2-2 [2] has MERV ratings, Minimum Efficiency Reporting Values. A MERV rating is the minimum efficiency for particles of potassium chloride measured for the clean filter and after three loadings of the ASHRAE standardized loading dust. ISO 16890 [3] produces three rating numbers for filters, ePM_1, $ePM_{2.5}$ and ePM_{10}, based on calculating the particle mass removed by the filter from two particle-size distributions typical of urban and rural locations. The subscripted numbers represent the approximate upper limits of particle diameter (in μm) for the ISO calculation procedure.

Filters do not always operate at the "rated capacity" set by the manufacturer. The efficiency of a filter is to some degree dependent on its flow rate. All the above-mentioned test standards ignore this fact, and require only that filter efficiency be measured at a single flow rate. If efficiency-vs-particle-size data were made available for low, medium, and high operating flow rates for a filter, efficiency values at other flow rates throughout the tested size range could be calculated by an interpolation algorithm.

The changing efficiency of a filter as it accumulates particles in actual operation is quite complicated, and not well defined in filtration literature. OAQ varies with season, day of the week, and hour of the day. Internal particle generation in many cases is time-dependent. These effects could be incorporated into a computer model to calculate particulate matter concentration, including its variation with time. In this discussion, we limit our model for particulate matter concentration to steady-state conditions, and accept filter efficiency as that measured at a single flow rate.

2. How PM_1, $PM_{2.5}$ and PM_{10} are defined and measured

Many journal articles and websites refer to PM_{10} as "the mass of particles less than 10 μm diameter". PM_1 and $PM_{2.5}$ are often similarly defined, as "…less than 2.5 μm…" and "…less than 1 μm…". These definitions are only approximations. PM_{10} is in fact the mass concentration of particles that would be

collected from an air flow by a sampler which has a size-selective device ahead of its filter medium. The size-selective inlet is designed to pass most particles with aerodynamic diameters less than 10 μm, and to capture most particles greater than 10 μm. The percentage passing, called inlet penetration, for an actual $PM_{2.5}$ inlet is shown in Figure 1.

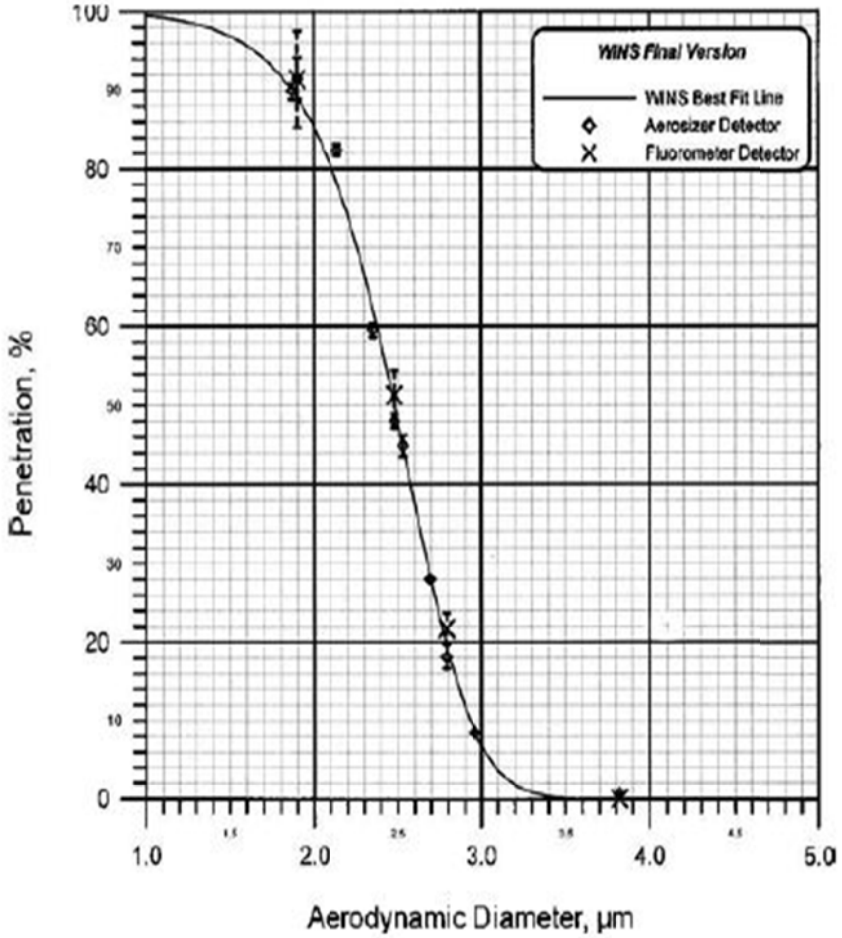

Figure 1. Particle penetration versus particle diameter for WINS $PM_{2.5}$ size-selective sampler inlet. (from reference [7]).

Note that the curve is tangent to the graph X-axes at penetration = 0% and 100%. It passes through diameter = 2.5 μm at penetration = 50 %, and transitions smoothly between these points. The curves for PM_1 and PM_{10} inlets are similar, but with 50 % values of 1 μm and 10 μm. The characteristics of PM_X sampler inlets are not, however, defined by the US Environmental Protection Agency and similar government agencies elsewhere as "ideal" for PM_1, $PM_{2.5}$ and PM_{10} by curves like Figure 1. The "ideal" curve for PM_{10} given in Ref. [8] is apparently a list of actual test data; and has irregularities which are not generally observed for impactors. The basic definition of $PM_{2.5}$ and PM_{10} in the United States, the Federal Reference Method (FRM) [8], is not data or a curve, but a set of drawings of the holder for the sampling media and the size-selective inlet that precedes it.

We have developed expressions which can be coded in computer programs for the PM_X inlet penetrations as functions of particle diameter. Our penetration function - equation (1) - fits the measured penetration data for PM_1 and $PM_{2.5}$ samplers quite well. It is a slight modification of an expression for the PM_X inlet penetrations given in Ref. [9]. Our modified expression is:

$$P_{in} = 100 \cdot \left[1 - \mathrm{K} \cdot \left(1 + \tanh\left(\mathrm{A} \cdot \left(\mathrm{B} \cdot d - \mathrm{PM_X}\right)\right)\right)\right] \qquad (1)$$

Here P_{in} is the inlet penetration in %; PMx is 1.0 or 2.5, depending on the PMx inlet simulated; and d is particle diameter in micrometers. For PM_1 we developed best-fit parameters for the penetration curve given in [10]. For $PM_{2.5}$, data were taken from Figure 1 above, from Ref.[7].

The values for K, A, and B are as follows for the PM_1 and $PM_{2.5}$ inlets:

- PM_1: K = 0.500; A = 6.3; B = 1.0; Diameter range: all
- $PM_{2.5}$: K = 0.52453; A = 2.08; B = 0.991 Diameter range: 0.0 – 3.256 μm

Equation (1) provided a rather poor fit to the PM_{10} inlet penetration curve. We smoothed the tabulated data in Ref. [8] using two polynomials, a cubic for the diameter range from 0.0 to 15 μm, and a parabola for the range from 15 to 16 μm, where the penetration reaches zero. These expressions for PM_{10} are:

- Over the diameter range 0 - 15 μm:

$$P_{in} = 96.6444 - 0.098681081 \cdot d - 0.56324808 \cdot d^2 + 0.01066718 \cdot d^3 \quad (2)$$

- Over the diameter range 15 - 16 μm:

$$P_{in} = 593.6 - 72.3 \cdot d + 2.20 \cdot d^2 \quad (3)$$

For all three inlets, penetration is 100 % for d below the lower end of the lower diameter range, and 0 % for d above the upper end of the upper range.

One thing to be kept in mind in all analyses related to PMx values is that they are measured or simulated only for the specific instantaneous particle-size distribution of Total Suspended Particulates (TSP) present, or its average over the sampling period. One should never speak of "a $PM_{2.5}$ dust" or "the PM_{10} efficiency of a filter" as values unconnected to the aerosol size distribution in the air of concern.

3. Particulate effects

HVAC systems designers are often faced with the problem of predicting the indoor air quality (IAQ) in buildings. The impetus for this is often the health of occupants, but it may be the effect of pollutants on materials or objects in the ventilated spaces [11]. In this discussion, we are concerned only with particulate pollution.

Many studies have been made of the impact of particulate pollution on human health. Indeed, concerns about outdoor pollution led to the establishment of PM-monitoring networks, and regulations based on PM data. The size-dependent toxicity of particles gave rise to the concepts of PM_1, $PM_{2.5}$ and PM_{10}. Many epidemiological studies provide correlations between PM_1, $PM_{2.5}$ and PM_{10} values and health effects.

Since humans spend much of their lives indoors, it is important to be able to determine the PMx levels that would be measured by a PMx sampler inside an occupied, ventilated space. This calculation might include the pattern of blower-driven flows for the spaces, leakages into and from the spaces, internal particle generation, particle deposition, air exchange between spaces, and the effects of filters in the system.

Characteristics of various system types are described in Refs. [12] through [15], and Ref. [13] and [16] provide equations for duct system design, including data on pressure drop in air distribution elements such as duct elbows. To calculate the flows in all system branches we need functions for the pressure drop dependence on air flow volume for all system elements. Both commercial and open-source computer program packages are available to carry out calculations of ventilation systems of considerable complexity.

In this discussion, we limit ourselves to one relatively simple configuration (Figure 2) and the choice of parameters usable in calculating its internal PMx values.

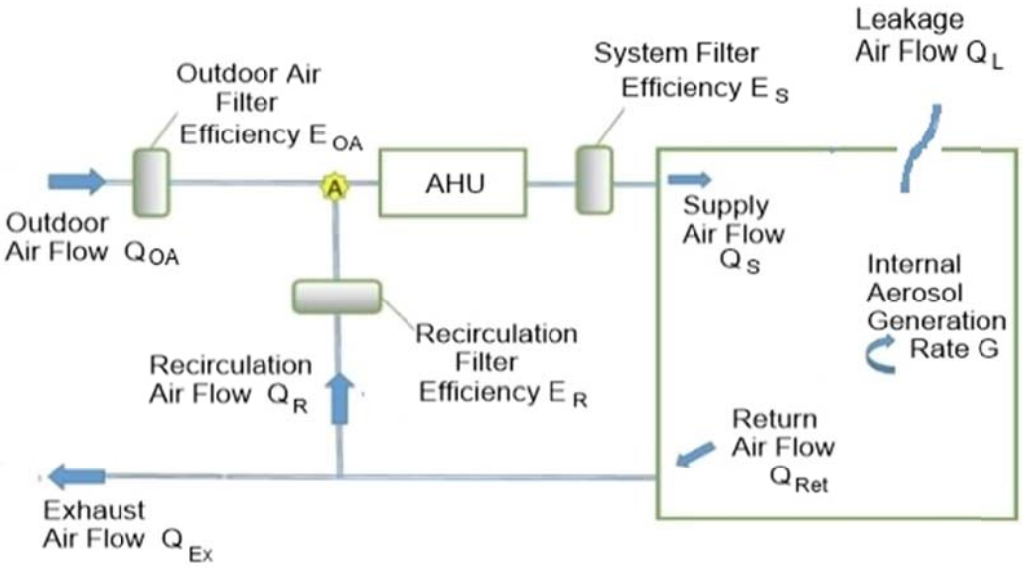

Figure 2. Schematic model of a ventilation system with recirculation and filtration for a single space.

4. Ambient particle size distributions

Many investigations have shown that outdoor aerosol distributions world-wide have three more or less distinct modes, each of which has an approximately log-normal distribution. The design of PM_1, $PM_{2.5}$ and PM_{10} size-selective inlets is not entirely arbitrary; they are intended to remove entire modes from TSP in a way that allows the mass in each mode to be determined. The entire outdoor mass size distribution in a locality can be described with fair accuracy if the three PMx values for the locality are known, and some added assumptions are made.

One must first accept some values for the density of each mode. In addition, one must have the geometric mean diameter and standard deviation of the modal log-normal distributions. The overall mass distribution as a function of particle size can then be calculated. If the mass distribution is obtained, one further assumption – that all particles are spheres – allows one to calculate the surface and count distributions. Alternatively, if we know the overall count distribution, and accept the assumed density and sphericity of particles and the modal parameters, we can calculate the details of the surface and mass distributions.

5. Calculating the effect of a filter on PMx

The log-normal function is one form of "probability density", which gives the relative numbers of particles of a specific particle size in an aerosol cloud. The most probable size has the geometric mean diameter d_{50}; probabilities for particles below and above this size have values given by the familiar bell-shaped "Gaussian" curve. The equation for this curve is:

$$f\left(d,\sigma_g,d_{50}\right)=\frac{1}{\ln\sigma_g}\exp\left[-\frac{\left(\ln d-\ln d_{50}\right)^2}{2\left(\ln\sigma_g\right)^2}\right] \quad (4)$$

Here d is the geometric mean diameter of a particle diameter range from d_1 to d_2, $(d_1 \cdot d_2)^{0.5}$, and σ_g the standard deviation of the modal distribution. The probability density of a multi-mode distribution is simply the sum of the probability densities of each mode, multiplied by the number-weighted fraction y for each mode. For three modes, the probability density is:

$$P(d) = y_A \cdot f(d, \sigma_{gA}, d_{50A}) + y_B \cdot f(d, \sigma_{gB}, d_{50B}) + y_C \cdot f(d, \sigma_{gC}, d_{50C}) \quad (5)$$

The sum of y values must always equal 1; in (5), $y_A + y_B + y_C = 1$. These y-values must be specified for the distribution of particle count, surface, or volume, depending on the parameter studied, or the method used obtain the distribution.

Equation (5) can be used for interior PMx calculations only if the density of individual particles is assumed to be the same throughout the entire size spectrum. This is a rather drastic simplification. The bulk of the mass in the PM_{10} mode, especially in rural locations, is often soil or sand particles with density about 2600 kg/m^3. The carbon and organic condensate particles which dominate in the $PM_{2.5}$ and PM_1 modes have densities nearer 1000 kg/m^3. Computer programs can accommodate such differences quite easily. We will therefore assume that the particles in a mode have uniform density, but that the density values for the modes are not necessarily equal. This means that for mass calculations, the masses in each narrow size range must be calculated separately for all modes present, and the values summed. The steps for a single mode follow here below.

If we take the area under the curve under the function given by (5) to represent the total number of particles in the aerosol sample, the number of particles present between two sizes d_1 and d_2 is given by two equations:

$$d = \frac{\ln d_2 - \ln d_1}{2} \quad (6)$$

and

$$\Delta N = P(d) \cdot (\ln d_2 - \ln d_1) \quad (7)$$

With the assumption that the particles in the mode are all spheres, with density ρ_m, the mass between d_1 and d_2 is:

$$\Delta m(d) = \rho_m \cdot \frac{\pi \cdot d^3}{6} \cdot P(d) \cdot (\ln d_2 - \ln d_1) \quad (8)$$

If this little mass packet is passed through a filter with an efficiency $E(d)$ for particles with diameter d, the mass $\Delta m_2(d)$ attributable to this mode between sizes d_1 and d_2 immediately downstream of the filter is:

$$\Delta m_2(d) = [1 - E(d)] \cdot \Delta m(d) \quad (9)$$

where $E(d)$ is the efficiency of the filter at size d.

The PMx concentration downstream of the filter including all particle sizes is:

$$PM_X = \sum_D \rho_m \cdot \frac{\pi \cdot d^3}{6} \cdot P(d) \cdot (\ln d_2 - \ln d_1) \cdot [1 - E(d)] \cdot P_{in}(d) \quad (10)$$

Here, $P_{in}(d)$ is given by Equation (1), (2) or (3), depending on which PMx is being calculated. Summation is performed over all D ranges. In effect, we have simulated what particles would arrive at the sampling filter of a PMX monitor.

Air filter efficiency as a function of particle size is generally available, at least for one “rated” operating velocity, and usually for the particle size range from about 300 nm to 10 µm. Existing international efficiency testing standards for general ventilation filters are limited to this range, but instrumentation needed to extend efficiency measurement to low nanometer particle sizes is increasingly available in testing laboratories. Filtration theory predicts, and tests confirm, that fibrous filters efficiency will generally increase below 100 nm, because of diffusion effects. Nevertheless, manufacturers need to extend filter testing to low nanometer sizes, so that indoor concentrations of nanoparticles can be calculated as accurately as possible. Such calculations require numerical values for efficiency over the entire range of interest, i.e. also below 300 nm size.

The European Committee for Standardization has recognized this need, and draft standards (EN ISO 21083-1 and -2) are being written defining test methods for measuring efficiencies on nanoparticles in two ranges, 20-500 nm and 3-30 nm.

6. Sources of data for ventilation system parameters

6.1. Outdoor size distributions

Particle size distributions outdoors have been measured and reported in numerous published papers, but the data are usually presented graphically, making them difficult to average or to incorporate into computer programs. Many distributions are presented in the cumulative, or "percentage greater than stated size" form, that is the integral of equation (4) from $d = 0$ to the stated value of d. It is possible, but difficult, to extract the modes from such plots. Often, the distribution data are based on measurements made with a cascade impactor having 4 or 5 stages. The parameters of mode probabilities that can be extracted from such measurements have very low accuracies. Our search for ambient mode data was limited to higher-resolution methods, such as the Optical Particle Counter (OPC), Tapered Element Oscillating Microbalance (TEOM), Differential Mobility Analyzer (DMA) and Scanning Mobility Particle Sizer (SMPS). Ref. [17] is a survey of these devices for measuring aerosol distributions. Each device has limitations on its useful size range. Table 1 gives the averaged modal parameters found in a group of these distribution references. The mode names shown are common in aerosol literature, though not very precisely defined. The last two rows of Table 1 give parameter values for the four modes suitable for indoor PM_x calculations.

Table 1. Averages of parameters measured for outdoor aerosol modes and recommended for simulations

Mode:		Nucleation		Aitken		Accumulation		Coarse	
Ref.	Location	d_{50}	σ	d_{50}	σ	d_{50}	σ	d_{50}	σ
20	Finland	11.7	1.71	37.3	1.78	151	1.60	-	-
20	Finland	13.8	1.63	42.5	1.71	152	1.54	-	-
21	Central Europe	-	-	67.4	1.68	265	1.49	-	-
22	United States	20.0	1.60	-	-	350	1.9	5000	2.2
23	Arctic Canada	-	-	-	-	300	2.2	10000	3.1
24	Europe	-	-	50.8	1.98	159	1.57	-	-
25	Pacific Ocean	21.8	1.16	36.0	1.19	155	1.18	-	-
26	Many	14.2	-	64.0	-	269	-	-	-
27	Beijing	-	-	-	-	305	1.67	6800	2.43
Recommended Values									
d_{50}, σ :		15	1.50	50	1.70	230	1.60	7300	2.60
ρ_m, kg/m^3:		1380		1000		1000		2660	

6.2. Outdoor PM concentrations

The World Health Organization provides a list of annual average $PM_{2.5}$ and PM_{10} values for 2971 cities world-wide [28]. The US EPA reports these data for thousands of urban and rural PM monitor sites [29]. Similar data is available for sites in other countries, for example [30]. There is essentially no data from government agencies for PM_1 concentrations. We list some values appearing in air-pollution literature in Table 2. The ratio (PM_1 / $PM_{2.5}$) for this list is reasonably constant. Multiplying its average value (0.75) times the local $PM_{2.5}$ might be used for PM_1 where no measured PM_1 is available.

Table 2. PM concentrations and $PM_1/PM_{2.5}$ ratios.

Ref.	Locale	Sampling Season, Days	Mean PM Values, μg/m3			Ratio,
			PM_1	$PM_{2.5}$	PM_{10}	$PM_1/PM_{2.5}$
31	urban, Vienna Austria	year 365	14.9	18.9	26.5	0.873
31	urban, Vienna Austria	year 365	14.7	18.8	29.1	0.782
31	suburban, Vienna Austria	year 365	17.5	21.1	31.0	0.829
32	urban, Athens Greece	year 365	18.5	23.7	51.3	0.781
32	urban, Athens Greece	year 365	20.1	29.3	52.2	0.686
32	university in hills near sea, Crete	year 365	10.3	17.9	32.5	0.575
33	urban, Taipei, Taiwan	spring 91	14.0	20.2	35.1	0.693
33	urban, Taipei, Taiwan	winter 90	9.7	12.7	26.4	0.764
33	urban, Taipei, Taiwan	spring 91	19.2	29.9	51.3	0.642
33	urban, Taipei, Taiwan	autumn 92	29.5	34.4	46.0	0.858
34	urban, Xi'an China	year 365	127.3	182.2*	-	0.699
35	urban, arid, Phoenix Arizona USA	spring 91	4.4	18.4	25.8	0.239
35	urban, arid, Phoenix Arizona USA	summer 92	5.9	8.4	81.6	0.702
35	urban, arid, Phoenix Arizona USA	autumn 92	9.9	14.2	57.8	0.697
36	urban rooftop, Chengdu China	spring 91	49	56	76	0.875
36	urban rooftop, Chengdu China	summer 92	40	43	49	0.930
36	urban rooftop, Chengdu China	autumn 92	54	56	60	0.964
36	urban rooftop, Chengdu China	winter 90	76	83	92	0.916
37	urban rooftop, normal year, Delhi	winter 90	204	236	338	0.864
37	urban rooftop, normal year, Delhi	summer 90	43	69	178	0.623
37	urban, monsoon season, Delhi	Aug./Sept 61	37	54	132	0.685
37	urban post-monsoon, Delhi	Oct./Nov 61	337	389	548	0.866
38	urban, highway traffic, Barcelona	July/Nov 150	17	25	38	0.680

6.3. Indoor particle generation

Extensive studies of indoor particle generation by industrial processes, such as welding, exist. The generation of particles in non-industrial situations, such as offices and concert halls, has received less attention, because the hazards are far less.

Aerosol generation by ink-jet and 3-D printers has been studied [39], [40], and there is abundant literature on the generation of particles and toxic gaseous contaminants by tobacco smoking. These studies have led to widespread prohibition of indoor smoking, but reference to the literature [41], may be necessary in some situations.

Nanoparticle generation in buildings is studied in [42] and [43]. The particulate progeny of radon and thoron are peculiarly hazardous because they are radioactive

6.4. System flows and leakage

The blower-driven flows Q_{OA} and Q_R in Figure 2 are of course determined by the heating and cooling loads of the space, and the number of occupants. Leakage into or out of the space or its ventilation ducts is dependent on the pressure differential between the space or ducts and outdoors, and can be measured the "blower-door" technique (Ref. [44]). Predicting leakage is difficult, but both new construction and remodeling usually seek to minimize leakage. Ref. [45] and [46] provide guidance on the effects of leakage-reduction. Ref. [47] provides actual measurements of leakage in buildings in several countries, with different wall constructions, where one may need to estimate leakage parameters for cases where no measurement of leakage is attempted.

7. Conclusions

Reasonable estimates of the total suspended particulates in ventilated spaces can be calculated, including the values of PM_1, $PM_{2.5}$ and PM_{10} mass concentrations that would be measured in these spaces by monitors designed to measure these indices outdoors. Such calculations allow evaluation of the impact of particle filters on the health and comfort of building occupants, and on potential damage to surfaces and objects in the ventilated space.

Some of the parameters needed for the calculation are well-defined and available for specific locations, but some are can only be assigned typical values. The calculations are complex, and well-organized user-friendly computer programs and databases are needed to make them useful to HVAC system designers.

The effect of filter loading with particles has not been quantified, and filter efficiencies-versus particle size at different operating flows are not usually available. This data must be provided if the cost and energy impact of filters are to be calculated.

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