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(Title)

Particulate Pollution Interactions with Indoor Surfaces: Measurement and Modelling for Risk Assessment and Contaminant Control.

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Synopsis.

In the urban environment, there is strong evidence that fine particulates associated with vehicular emissions are linked with respiratory problems and an increase in mortality. The population sector most at risk is the elderly who spend much of their time indoors; consequently, the infiltration of these particles and their subsequent behaviour indoors is of primary concern.

The deposition of aerosol particles in the respiratory system and on indoor surfaces is a process governed by particle size; in addition to providing risk assessment data, an understanding of the interaction of particles with indoor surfaces can be applied to the design of systems for enhancing indoor aerosol deposition and thus inhibiting inhalation exposure. This paper describes experiments using tracer-labelled aerosols, in a range of monodisperse size distributions representative of real particulate pollutants, to study aerosol deposition on surfaces with representative roughness. Some preliminary data, exhibiting electrostatically-enhanced aerosol deposition are also presented. In order to make these data widely accessible, the experimental results are used to aid in the development of a CFD code by providing validation data. Simulations are described for a room-sized enclosure with representative indoor surfaces, to illustrate the influence of internal building surface characteristics on indoor aerosol concentration modification.

1. Introduction.

Air pollution studies in cities of the United States and in London have demonstrated a strong association between urban aerosol concentrations (largely attributed to vehicular emissions) and excess numbers of deaths from respiratory diseases. Since excess deaths are most clearly seen in the elderly sector of the population, the U.K. Department of the Environment's Expert Panel on Air Quality Standards conclude that particulate pollution episodes are most likely to exert their effects on mortality by determining the time of death of those rendered susceptible to pre-existing disease; however the possibility also exists that prolonged exposure to air pollution may contribute to disease development (1).

Since the elderly sector of the population spends much of its time indoors, and often in naturally-ventilated dwellings, indoor inhalation exposure to particulates of outdoor origin is of primary concern. Ingressed aerosol is either removed from indoor air through ventilation or is deposited on internal building surfaces; aerosol deposition rate measurements (2) have shown that, particularly at low air exchange rates, particulate deposition is an important modifier of inhalation exposure. Aerosol deposition, both inside the respiratory system and on indoor surfaces, is a particle size-dependent phenomenon.

The motion of the largest particles (those greater than 1 μm), which lodge in the upper airways, is gravitationally-dominated; the inhalation of these particles could therefore be inhibited by the promotion of turbulent deposition and impaction on indoor surfaces.

Particles smaller than 1μm penetrate more deeply into the lung (3); particles at the upper end of this range (0.1 -1μm), are neither strongly influenced by diffusive effects or gravity. It is expected, therefore, that external forces would play a role in enhancing the deposition of these particles on indoor surfaces, thereby reducing their inhalation potential; in the indoor environment, such forces commonly arise, for example, from electronic visual display units which generate electrostatic gradients. Particles smaller than 0.1 μm are highly susceptible to alveolar deposition, due to Brownian diffusion; since these particles behave essentially like a gas, their inhalation potential is largely governed by air-exchange.

To provide risk assessment data for airborne particulate exposure and to examine whether the forced enhancement of aerosol deposition on indoor surfaces might be an effective means of inhalation dose modification, a detailed understanding of the mechanisms controlling aerosol deposition is required. Few direct measurements of deposited aerosol particle masses on internal building surfaces exist since such studies require sensitive multi-elemental analytical techniques which are not widely available; the techniques employed at Imperial College for measuring aerosol deposition rates to internal building surfaces are described in the next section. In order to make these data widely accessible and limit the necessity for further costly and labour-intensive measurements, the experimental results are used to aid in the development of a CFD code by providing validation data. The ultimate objective is to develop a code which predicts accurate particle/surface interactions for a range of ventilation conditions, surface features and particulate characteristics.

Following the description, in the next section of this paper, of the techniques used for measuring aerosol deposition to surfaces, the main features of the computational model are summarised. The current status of model development is then illustrated, by comparing predicted aerosol-surface interactions with the measured values.

2. Experimental.

2.1 Aerosol generation and labelling.

The techniques used for generating, dispersing and detecting tracer labelled particles have been previously described (4). Porous silica particles, available in a variety of supra-micrometre monodisperse size distributions (with mass median aerodynamic diameters of 2.5 μm, 4.5 μm and 5.4 μm, respectively), are agitated in a tracer salt solution so that tracer ions become bound to the particles' surfaces. The labelled particles are dispersed using a rotating brush aerosol generator. Sub-micrometre tracer particles, with a mass median aerodynamic diameter of 0.7 μm, are generated by atomisation and subsequent evaporation of a tracer salt.

Salts containing the rare earth elements dysprosium and indium are used as tracers in this work. Both exist naturally in a stable state but become unstable (i.e. radioactive) when bombarded with neutrons. Subsequent to a period of aerosol deposition in a test room/chamber, neutron irradiation of tracer aerosol-bearing materials (such as air filter papers or samples of domestic furnishing materials) in a nuclear reactor, followed by gamma-spectrometry, allows a quantitative determination of the aerosol mass present. Since dysprosium and indium occur naturally in low concentrations, the possibility of analytical interferences from particle-bearing media are minimised.

2.2 Aerosol deposition measurements in a test chamber.

The sensitive aerosol detection techniques summarised above have been employed to study aerosol deposition in the Imperial College test chamber. The chamber is an aluminium cube of side 2m, fitted with externally-accessible air sampling ports. An internal fan, situated on the central axis and pointing towards the floor, provides a source of inhomogeneous turbulence, in the range 22-43%. After a period of aerosol injection and deposition, during which air sampling is carried out, filter papers attached in regular arrays to the internal surfaces of the test chamber are analysed. Since the initial aerosol concentration in the chamber can be calculated by air sample analysis, the proportion of aerosol particles deposited on each internal chamber surface can be inferred from the surface filters. The integrity of this approach has been proven by aerosol mass balance tests; the correction factors (which includes the aerosol particle mass deposited on the fan blades) involved in the mass balance calculations are discussed elsewhere (5).

Rough materials can be mounted over the smooth internal aluminium surfaces of the test chamber so that the effect of surface roughness on aerosol deposition can be studied. Direct sampling of the rough surface, subsequent to the aerosol deposition period, is avoided in order to minimise particle losses and aerosol deposition enhancement to the rough surface is inferred by comparison with the smooth surface case.

Using the four particle size distributions available, the relative proportions of aerosol particle mass deposited on the walls, ceiling and floor of the unlined test chamber were determined. The chamber walls were then lined successively with wallpaper, short-pile carpet, and astroturf and the enhancement in deposited aerosol particle mass was measured. The roughness of the surfaces was quantified by friction velocity measurements; using a hot-wire anemometer in a small-wind tunnel, the variation in air velocity with height above each surface was measured. The results, for two particle sizes, and smooth and rough chamber surfaces, are compared with model predictions in section 4.

In the indoor environment, persistent soiling of computer monitors and television screens provides evidence of the preferential deposition of aerosol particles on electrostatic surfaces. The deposition of charged aerosol particles on charged surfaces of opposite sign is easily understood; the mechanism by which uncharged particles deposit can be explained by considering that the charged surface induces an "image charge" of opposite sign on the particles, promoting attraction towards the surface and subsequent deposition (6). Using the test chamber, preliminary measurements were carried out whereby the deposition velocity (i.e. the aerosol particle mass deposited on a surface of unit area in unit time, divided by the particle mass concentration in the core of the enclosure) of 0.7 μm and 2.5 μm particles to vertical polythene sheeting was determined. In these measurements, the air turbulence in the test chamber was minimised, so that electrophoresis would be the dominant influence on aerosol transport. The results are shown in section 4.

3. Computational.

3.1 Introduction.

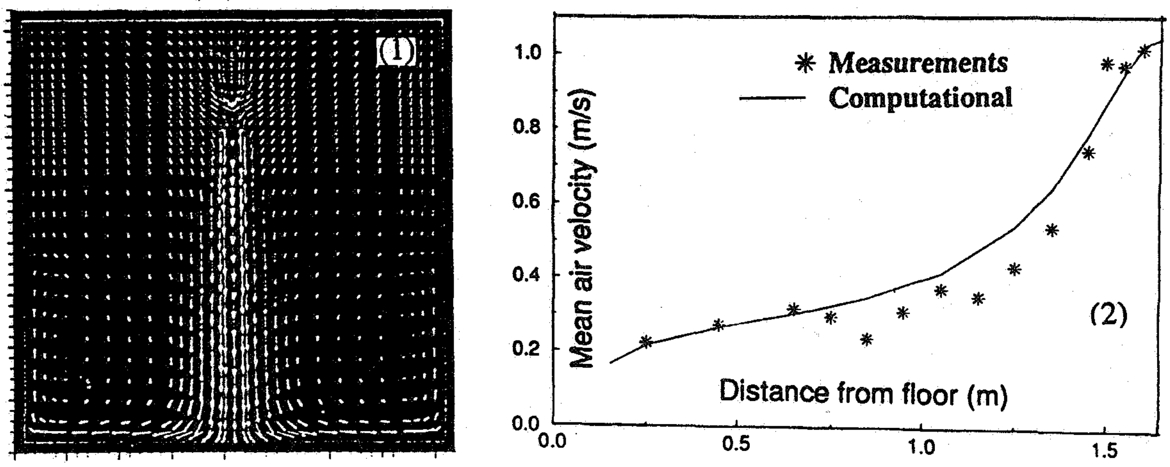
While a large number of researchers have studied the physical mechanism underlying particle deposition on surfaces, the research has largely been confined to the viscous sub-layer close to a surface, and deposition has been considered as a process isolated from the main turbulent flow field. The mechanism by which particles are transported to the boundary layer by the turbulent dispersion of particles in the main stream is an important and frequently overlooked phenomenon and to address this, an analytical method called the "Prediction of Evolving Probability" (PEP) model, in which the evolution in time of the particles' velocity probability density is derived from first principles, has been developed (7). This method considers all possible realisations of the turbulent flow field to obtain the evolution of the particles' velocity probability density function, with the result that the need for time-consuming stochastic trials is eliminated.

The present study adopts the PEP method to model the particle-turbulence interaction, along with the more conventional free-flight model to simulate the whole deposition process. The main features of the computational methods are summarised below.

3.2 Modelling the gas-phase.

The CINAR CFD code, which solves the governing equations in a Cartesian co-ordinate framework, has been applied to predict flows in large multi-burner industrial furnaces and other complex configurations. The standard two-equation 'k-ε' closure is employed to model the gas-phase turbulence and a finite difference/finite volume technique is used to solve the Navier-Stokes equations on a conventional staggered grid.

Using the mean air velocity value at the centre of the test chamber (measured using a Dantec 5410 hot film anemometer) to define the momentum source term due to the mixing fan, the CINAR code was used to simulate the gas-turbulence interaction, as shown in Figure 1. Figure 2 shows the computed mean velocity profile above the floor; good agreement with the measured velocity values is observed.



Figures 1 & 2. CFD simulation of the airflow generated by the use of the mixing fan in the test chamber (1) and comparison of experimental and computational air velocity values on the central axis of the chamber (2).

3.3 Modelling the particulate phase: the PEP model.

The particulate phase is simulated in a Lagrangian frame and the contributions of the particulate phase to the source terms of the gas-phase equations are evaluated according to the scheme proposed by Migdal and Agosta (8). The effects of the particle-turbulence interaction on the motion of the particulate phase are accommodated by the PEP model, assuming that the joint gas-particle velocity distribution is Gaussian. The evolution equation for the probability density function (pdf) of an ensemble particle velocity may be derived as follows (9) :

$$C_o[C_1+C_2v_p]\,P_{v_p} + P_t + C_oC_1\,P = 0 \qquad [1]$$

where

$$C_o = (\rho_g\, A_p\, C_D\, /\, 2\, m_p)\,.\; u_g - u_p\ ,$$

$$C_1 = \bar{u}_g - \frac{\rho_{gp}\sigma_g}{\sigma_p}\bar{u}_p\ ,$$

$$C_2 = \frac{\rho_{gp}\sigma_g}{\sigma_p} - 1\ .$$

A_p and m_p are, respectively, the particle mass and surface area. C_D is the drag coefficient and ρ_g is the gas velocity. P denotes the pdf $P(v_p,t)$, v_p being the fluctuating particle velocity and t, the time; the subscripts v_p and t denote the partial derivatives with respect to these variables. u_g and u_p denote the mean values of gas and particle velocity in real space. σ_g and σ_p are the rms values of the gas-phase and particle-phase velocity fluctuations, respectively, and ρ_{gp} is the correlation coefficient for the gas-particle velocity fluctuations. The solution of equation [1] can be obtained using the method of characteristics. The discrete time step for the particle tracking is estimated as the minimum of either the particles' dynamic relaxation time or the time step based on the local cell dimensions.

3.4 The deposition model.

The deposition model used in the present simulation is based on the "stopping distance" concept and is similar to the commonly-used free-flight model (10). The steps in the deposition model are as follows: all cells are successively searched until a cell containing a particle is located. That particle is then traced through the flow field until it either exits the domain or arrives within a pre-calculated distance (which depends on the friction velocity) of the wall. If the particle leaves the domain, calculations for this particle are terminated and a new particle is initialised. Alternatively, if the particle reaches the edge of the sublayer, the velocity component normal to the wall is calculated. This velocity is used as a free flight velocity to determine whether the particle will penetrate through the sublayer and if the particle's incoming velocity is less than or equal to a critical velocity, the particle is assumed to be deposited. The critical velocity for each particle is calculated by incorporating a particle bounce model (11). In this, the energy loss mechanisms are theoretically modelled by considering the relationship between contact deformation mechanics and contact surface energy.

In the case of a rough wall surface, an additional deposition mechanism is provided and the deposition model must be modified. The mean velocity profile due to the roughness of the surface is shifted, using the correlation of Im and Ahluwalia (12), so that the zero velocity level is no longer at the wall, but some height above it. It is assumed that if a particle reaches a distance equivalent to the mean plus one standard deviation of the roughness elements with respect to the zero velocity level, it will be captured by the wall.

Computations were carried out for two monodisperse particle size distributions: 2.5 μm and 4.5 μm particles. In successive computations, the roughness heights of the chamber wall surfaces were adjusted to correspond to those of aluminium, wallpaper, carpet and astroturf.

4. Results.

Figure 3 shows the predicted proportion of 2.5μm and 4.5μm particles which deposit on the test chamber surfaces, compared with the experimental results. The model predicts that a significantly greater proportion of the particle mass is deposited on the floor than on the walls of the test chamber, in agreement with the measurements. These findings seem reasonable since the direction of airflow in the chamber, as dictated by the presence of the fan, is vertically downwards. In addition, the actual percentage of particles deposited on the floor, as predicted by the model, is greater in the case of the 4.5 μm particles than the 2.5 μm particles; this result is also in agreement with the experimental observations and confirms the anticipated gravitational influence in the larger particle size range.

Figure 4 shows the measured and modelled proportions of aerosol particle mass deposited on the rough test chamber walls. It can be seen that the measured values exceed those predicted by about a factor of two, and that the model is less sensitive to changes in surface roughness. While the interaction of the particles with the rough surface is modelled within the sub-layer, the height of the roughness elements (11mm in the case of astroturf) is such that it is likely that they penetrate into the core of the flow field, causing inertial impaction of particles. The difficulty in incorporating this process in the model is thought to be largely responsible for the discrepancy between the predicted and measured values.

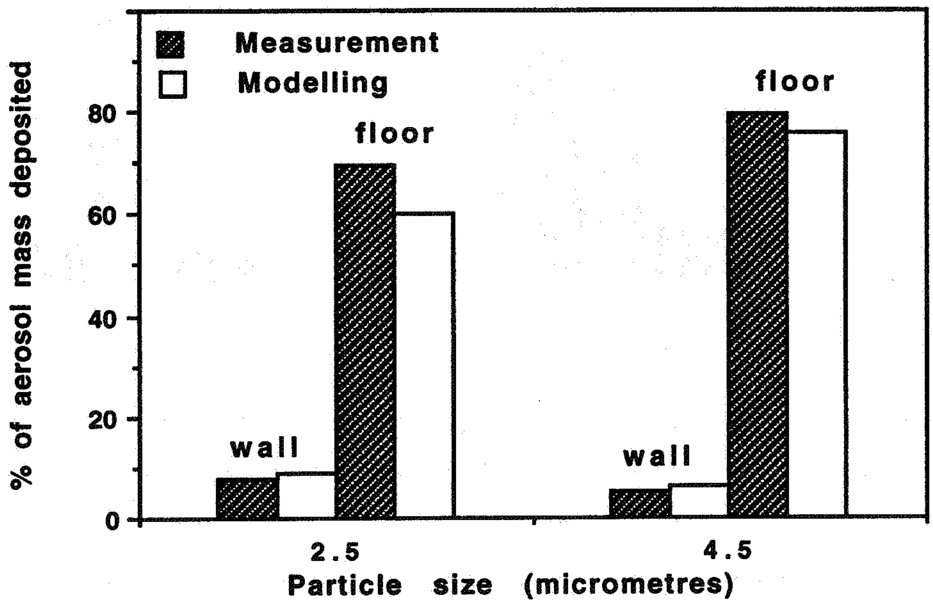


Figure 3. The proportion of aerosol particle mass deposited on the floor and one wall of the aluminium test chamber: measured and predicted values for 2.5 μm and 4.5 μm particles.

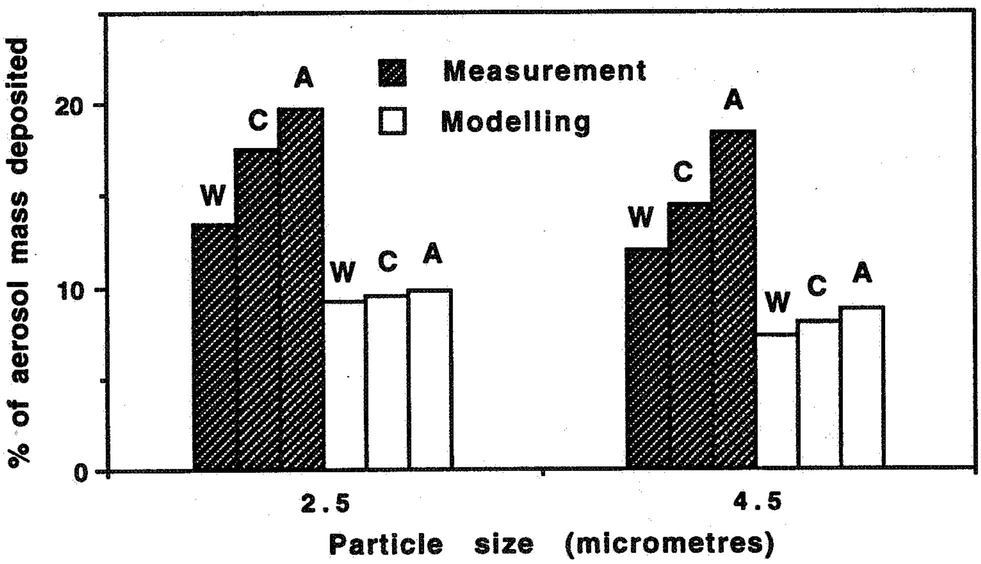


Figure 4. The proportion of aerosol particle mass deposited on a test chamber wall lined successively with wallpaper (W), carpet (C) and astroturf (A): measured and predicted values for 2.5μm and 4.5μm particles.

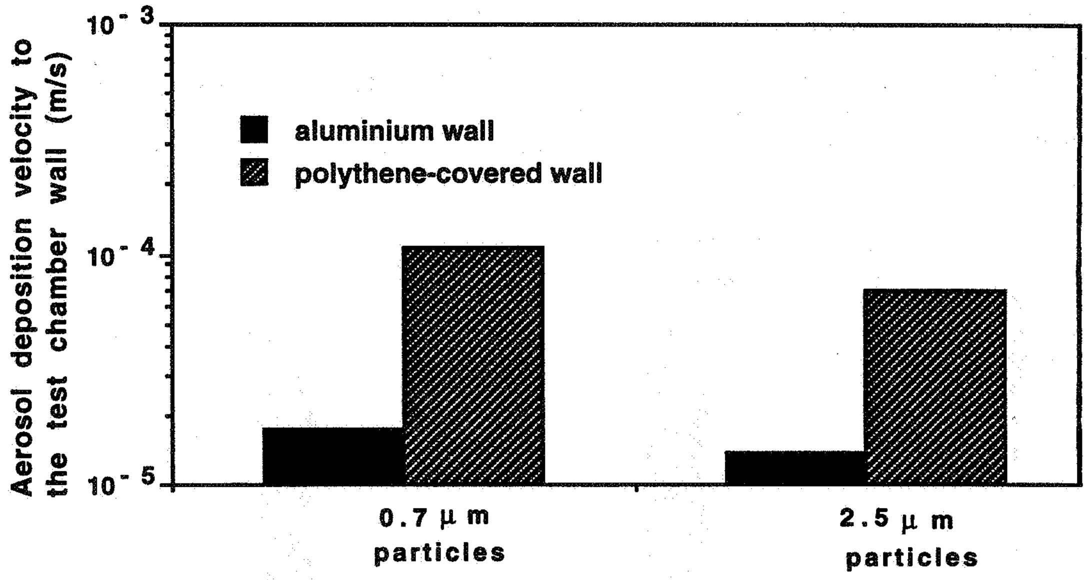


Figure 5. Measured aerosol deposition velocity to the test chamber wall, for 0.7 μm and 2.5 μm particles; comparison of a conducting (aluminium) and an electrostatic (polythene) wall surface.

The observed enhancement in aerosol deposition velocity to the wall of the test chamber, when that wall is lined with an electrostatic material, is shown in Figure 5. It can be seen that the effect is most pronounced for the 0.7 μm particles, since they are less strongly influenced by gravity than the 2.5 μm particles. The measured deposition velocity of the 2.5 μm particles to the aluminium floor of the chamber was 2.5x 10^{-4} ms^{-1}, which was significantly greater than that measured to the polythene-covered wall. This finding confirms earlier research (13) which concluded that electrostatic effects were most influential for particles less than 1μm in diameter.

5. Conclusion.

It has been shown that the deposition of particles on indoor surfaces, which is an important modifier of indoor inhalation dose for particulate pollutants, is strongly influenced by the characteristics of those surfaces. Results suggest that indoor air pollution control strategies should exploit the sensitivity of supra-micrometre particle transport to turbulence and impaction effects and the preferential deposition of sub-micrometre particles to electrostatic surfaces.

A computational model has been shown to accurately predict the interaction of particles with smooth surfaces in a room-sized enclosure. The prediction of aerosol deposition to rough surfaces is complicated by the process of inertial impaction of particles on roughness elements. Future work will aim to incorporate this process in the model; simulation of the deposition of particles to electrostatic surfaces will also be carried out.

Acknowledgement.

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