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Particle Deposition in Human Respiratory Tract: Effect of Water-Soluble Fraction

Suresh K. Varghese[1*], S. Gangamma[2]

[1] *Department of Civil Engineering, National Institute of Technology Karnataka, Surathkal, Karnataka, India, 575 025.*

[2] *Department of Chemical Engineering, National Institute of Technology Karnataka, Surathkal, Karnataka, India, 575 025.*

Abstract

In the nearly saturated human respiratory tract, the presence of water-soluble substances in inhaled aerosols can cause change in the size distribution of particles. This consequently alters the lung deposition profiles of the inhaled airborne particles. The magnitude of particle deposition in the lung is affected by the soluble component present in the particle. This is estimated by a numerical model. The model solves the condensation growth equation to determine the size evolution of respirable particles within the human respiratory tract. The water uptake by the particles in the respiratory tract results in change of size and density of the particles, and these changes are incorporated for estimating the particle deposition efficiency. The model results are compared with experimental results of sodium chloride particles. The model reproduces the major features of the experimental data. The simulations indicate that the particle can grow up to two times or more of its original size due to water uptake, depending on the quantity of the soluble matter it carries and thus can have significant effect on particle deposition efficiency. The study investigated the effect of soluble compounds in estimating total and regional lung dose of ambient particulate matter measured in Mumbai, India. The particle mass size distribution and composition considered for the ambient particulate matter, the variation in the total mass dose due to the growth was modest. But, the regional lung dose was significantly affected by the hygroscopic growth.

Keywords: Atmospheric particulate matter; Lung deposition; Hygroscopic growth.

[*] Corresponding author. Tel: +91-0824-2474604, Fax: +91-0824-2474033

E-*mail address*: varghesesk@gmail.com

INTRODUCTION

Inhalation is the predominant exposure pathway to anthropogenic airborne pollutants. Knowledge of the behavior and fate of the inhaled aerosols in the human respiratory system have important applications in the field of inhalation toxicology. Deposition site and dose of inhaled particles in the lungs are important for assessing the toxic effects of inhaled particulate matter. Similarly, in aerosol therapy, the efficiency of therapeutic method is related to the dose delivered to the target rather than to the amount inhaled. The dose and the site of the deposition of aerosols are determined by various mechanisms of particle deposition and these mechanisms are strong functions of aerosol size. Therefore, changes in the size of the particles in the respiratory tract can influence the deposition pattern of the particles in the lungs. The uptake of water vapor by soluble components of airborne particles is a possible pathway for changes in the particle size distribution. Ambient conditions may not meet the conditions of super saturation required for the condensation process on the insoluble particles. But, the presence of water-soluble or hygroscopic compounds in the atmospheric aerosols can modify the size distribution at high humidity levels in the respiratory tract. The near-saturation conditions in the respiratory tract promote vapor transport between air in the airway lumen and hygroscopic particles.

Actually, many pharmaceutical formulations used in aerosol therapy contain hygroscopic substances which modify particle deposition characteristics (Gonda, 1992). The deposition profile along the respiratory tract is altered and individuals exposed to such airborne matter may be subjected to enhanced or reduced deposition rates compared to those of insoluble particles (Martonen *et al.*, 1982). The effect of water uptake on deposition efficiency of soluble aerosols in the human respiratory system has been investigated experimentally and theoretically (Blanchard and Willeke, 1984; Broday and Georgopolous, 2001). However, in the previous studies, the effects were estimated for a single hygroscopic compound, or simple combinations of them. The growth and deposition of the ambient particulate matter which carries a multitude of compounds has not been investigated.

The present study focuses on the commensurate effects of water vapor uptake and deposition of ambient atmospheric particulate in human respiratory tract. Composition and size distributions of ambient atmospheric aerosols are taken from a field measurement in Mumbai, India (Venkataraman *et al.*, 2002). In the following sections, the methodology and model equations are explained. The results of the simulations indicate that hygroscopicity is a critical factor affecting the deposition of inhaled ambient atmospheric particles.

METHODOLOGY

In this article, single particle approach is followed to address the effect of water uptake by

inhaled particles on the particle deposition. This means that the particle growth and deposition efficiency is estimated for the individual particle size bins and then integrated over the mass size distribution to estimate the lung dose. An aerosol condensation growth equation (Fuchs, 1959) is solved by using a geometric model (Weibel, 1963) of the human respiratory system. A one-dimensional Lagrangian (Schum and Yeh, 1980) formulation is used to calculate the simultaneous size-resolved growth and deposition of aerosols as they move along successive lung airways, starting at the nares region. The aerosol is assumed to be a part of an air parcel moving at mean velocity of parcel and dividing through the airway generations. The dispersion is ignored in the model formulation. First, the change in diameter of the particle of each size bin in the parcel as the particle traversed through the generations of the lung airway is calculated. The average modified diameter in each airway is used for estimating the size-resolved deposition efficiency. The methods adopted for these calculations are described in the following sections.

The condensation growth of a single particle and its change in the diameter and density is addressed. Then, formulae are introduced for calculating the deposition efficiencies of inhaled particles in the nasopharyngeal (NP) region, adding the effects of hygroscopicity on the particle size and density. Finally, the method of calculation of deposition efficiency in the rest of the airway sections and effect of condensation growth are described.

Condensational growth of single particle

Single particle condensation growth, describing particle size and mass change that result from vapor diffusion to a single spherical droplet, is applied to obtain the growth of atmospheric particles (Fuchs, 1959; Davies, 1978). The topic is extensively studied and therefore only a brief overview of single particle growth is provided here. The governing equation for mass transfer of water vapor to a droplet can be expressed in continuum regime as,

$$\frac{dm}{dt} = 2\pi D_p D_v (c_\infty - c_{eq}) \tag{1}$$

where, m is the droplet mass, D_p its diameter, D_v the water vapor diffusivity (temperature dependent), C_∞, concentration of water vapor, C_{eq} the equilibrium water vapor concentration of the droplet. Using ideal gas law and mass balance, the Eqn. (1) can be modified as

$$D_p \frac{dD_p}{dt} = \frac{4D_v M_w}{R\rho_p}\left[\frac{P_w(T_a)}{T_a} - \frac{P_w(D_p, T_d)}{T_d}\right] \tag{2}$$

Where, M_w is the molecular weight of water, R universal gas constant, ρ_p is the particle density, D_v diffusivity of vapor molecules, T_a the air temperature far from the droplet, T_d the droplet temperature (assumed to be uniform), P_a the partial vapor pressures far from the particle and P_w (D_p, T_d) is the equilibrium water vapor pressure of the particle.

The driving force in Eqn. (2) is a gradient in the water vapor concentration between the equilibrium concentration near the particle and away from the particle. The equilibrium water vapor pressure over flat surface is taken from Yaws (1999). The droplet equilibrium vapor pressure is calculated by accounting for the effect for curved surface on equilibrium vapor pressure (Kelvin effect), and solute effect of hygroscopic fraction with an insoluble core (Pruppacher and Klett, 1978).

$$\ln\left(\frac{P_w(D_p)}{P_w}\right) = \frac{A}{D_p} - \frac{B}{\left(D_p^3 - D_u^3\right)} \qquad 2(a)$$

Where,

$$A = \frac{4M_w\sigma}{RT\rho_w} \qquad 2(b)$$

$$B = \frac{6S_nM_w}{\pi\rho_w} \qquad 2(c)$$

σ is the droplet surface tension, ρ_w water density, S_n moles of solute dissolved in the droplet and D_u is the equivalent diameter of the insoluble fraction of the particle. Approximations for dilute solutions are used in the calculation of solute effect. The approximation of ideal solution, pertinent to Rault's law, deviates significantly from actual data on the behavior of concentrated solutions (Pruppacher and Klett, 1978). This is of concern for the solution droplets found in the initial growth phase of crystalline particles and saturated solution droplets.

Eqn. (2) neglects the non-continuum effects, that may induce error in the small droplets (Knudsen number >> 1) growth estimation. The equation is corrected by using modification of diffusion coefficient (Pruppacher and Klett, 1978; Hinds, 1999). Accommodation coefficient value is taken as 1 for the calculations. The equilibrium vapor pressure in Eqn. (2) corresponds to the droplet surface, which is a function of temperature. During water condensation, heat is released at the droplet surface and the droplet surface temperature is expected to be higher than the ambient temperature.

The energy balance at the droplet surface, assuming instant energy transfer inside the droplet,

gives (Seinfeld and Pandis, 1998),

$$2\pi D_p k_a'(T_a - T_d) = -Le\left(\frac{dm}{dt}\right) \tag{3}$$

where, k_a' is the thermal conductivity of air (corrected for non-continuum effects) and Le is the heat released by unit mass condensation (evaporation) at constant temperature. Since the particle Reynolds number and Schmidt's number of water vapor for typical lung conditions are smaller, the convective corrections for vapor and heat transport are neglected in the calculation. However, the upper airways where turbulence normally prevails due to structural elements that obstruct the airflow promote turbulent eddies even at moderate Reynolds numbers. Under such conditions the convective flux may play a major role in supplying fresh vapor to the particles (Broday and Georgopolous, 2001).

The uptake of water and growth of the particle can change the density of particles. Consequently, the particle deposition efficiency through impaction and sedimentation also may change. The diameter of the particle at a given time can be calculated by Eqns. (2) and (3). The corresponding density change is then computed by using the following formulae from Martonen (1982).

$$\rho(t) = \left[\frac{D_p(0)}{D_p(t)}\right]^3 \left(\rho(0) - \rho_w\right) + \rho_w \tag{4}$$

where ρ_w density of water and $D_p(0)$ and $\rho(0)$ are the initial diameter and density of the particle, respectively.

Growth and deposition in nasopharyngeal region

Hygroscopic growth rates of inhaled aerosol particles are a function of airstream properties, aerosol physicochemical characteristics, and airway morphology (i.e., cross-sectional area affects the flow velocity, which in turn affects the residence time of the particle). The airways of the human head and throat are complex structures and are difficult to describe mathematically. In this article, two cylindrical compartments are used for describing the sections from the nares to the trachea (Overton, 2001). The total volume of the two compartments is adjusted equivalent to average volume of extra-thoracic of Indian women (Varghese *et al.*, 2005). The volume-to-surface-area ratio is used for the calculation of the cross-sectional area based on Subramaniam et al. (1998). Due to the complex nature of the NP region, deposition studies typically rely on

empirical or logistic models. The deposition model employed herein is that advocated by Martonen and Zhang (1992).

$$\eta = \left(1 - \exp\left(-39.9 Q^{-0.14} D^{0.599}\right)\right) + \left(1 + \exp(12.39 - 2.92 \log(\rho D_p{}^2 Q))^{-1}\right) \quad (5)$$

where, η is the deposition efficiency, Q volumetric flow rate, (cm^3/s), D diffusion coefficient of the particle (cm^2/s), ρ particle density (gm/cm^3), and D_p particle diameter (μm).

To address the effects of hygroscopicity on deposition, the growth formulae from Eqns. (2) and (3) are incorporated into the aforementioned deposition efficiency Eqn. (5). Hygroscopicity will affect the diameter and density of an inhaled particle as a function of its residence time in the NP region. Thus, the deposition efficiency equation can be treated as a time-dependent function.

Particle deposition in lung

Various deterministic models of particle deposition in the respiratory tract have been formulated to calculate the deposition of inhaled particles in humans and animals. The models range from empirical models that do not incorporate lung geometry (Rudolf, 1986) to mathematically complex, many-path (multiple-path) models and stochastic models (Anjilvel and Asgharian, 1995; Asgharian *et al.*, 2001). Ideally, an anatomical model of the respiratory tract should simulate all of the paths (each being unique) from the upper respiratory tract (URT) entrance to the most distal airspaces. But in this article, anatomical characteristics of the respiratory tract are represented by a single path, symmetric, or an identical-path anatomical model. The use of identical-path type anatomical models is well established for respiratory tract particle deposition. Thus, for a given generation or model segment, the dimensions of one airway, or airspace, and the number of airways, or airspaces, in the generation completely define the characteristics of the given anatomical model generation. As a consequence, only one path needs to be considered. This simplifies the respiratory tract dosimetry modeling of particulate matter. Even though simplified, this model is useful for obtaining average regional and overall deposition of the particles in the lung (Asgharian *et al.*, 2001).

The lung geometry follows Weibel's symmetric dichotomous model (Weibel, 1963), scaled to specific functional residual volume and tidal volume. The scaling procedure from a total lung capacity to the functional volume is described in Schum and Yeh (1980). For any given FRC (functional residual volume) and TV (tidal volume) the diameters of the conducting airways and lengths and diameters of distal airways are calculated. Unless specified, the respiratory functions were assumed to reflect the respiratory physiology of an Indian woman involved in household activities (Varghese *et al.*, 2005). This corresponds to a tidal volume of 820 cm^3 and a breathing

frequency of 22 per min. These values represent the average of experimental data on medium aged adult women involved in various household activities. The household activities include cooking, dish washing, scrubbing, vacuuming, etc. These respiratory functional properties are similar to an adult female involved in slow walking (Adams, 1993).

Aerosol deposition calculations are estimated by considering major mechanisms as diffusion (Ingham, 1975; Landhal, 1963), sedimentation (Beeckmans, 1965) and inertial impaction (Yeh, 1974). For expiration phase of calculations, the model assumes that there is no impaction. The fraction of material deposited during inspiration at each generation n, is calculated as the concentration of aerosol in generation n^{-1} times the fraction of tidal air penetrating to n, times the overall removal efficiency in n. The fraction deposited and concentration at each generation is computed for specified particle properties at a constant flow rate determined by tidal volume and breathing frequency. For the last generation to which a given tidal volume penetrates, the efficiencies are calculated by means of mean residence time of the tidal air in that generation, rather than airstream velocities. The fraction deposited in lung region is corrected for particles deposited in nasopharyngeal region.

The model parameters

The diameter and density of the particle at each generation is obtained by integrating simultaneously the growth Eqns. (2), (3) and (4) by using ODE solver DVODE (Hindmarsh, 1983). A geometric average of the particle size and density is calculated for each generation in the respiratory system. The modified average particle properties are used for calculating the deposition efficiency for each generation. The temperature and relative humidity (RH) profiles along the upper respiratory tract are necessary inputs for estimating particle growth and deposition in the human airways. However, such data on the measurements of temperature and humidity profiles in respiratory tract are non-existent. The profiles are usually estimated using models, rather than from direct measurement; and predicted profiles show large variability in the NP region among individuals, owing to differing physiological characteristics and environmental conditions and breathing modes (Ferron *et al.*, 1985).

In this article, based on model calculations (Naftali, 1998), constant temperature and RH profiles at 37 ˚C and 90% were assumed for the extra thoracic region. Constant temperature and RH were assumed to be 37 ˚C and 99.5%, respectively, along the lung airways. The validity of the constant humidity profile along lung airways depends on whether water condensation has significant effect on drying the inhaled air. Typical hygroscopic aerosol growth requires negligible drying of the inhaled air for reasonable residential, occupational, or ambient temperature and humidity conditions. Therefore, the coupling of particle growth with air dehumidification within the airways is unimportant for normal exposure to ambient pollutants

(Broday and Georgopolous, 2001).

Ambient aerosol properties

The ambient aerosol size distribution and composition parameters were adopted from a field measurement in Mumbai, India (Venkataraman *et al.*, 2002). The measurements were carried out in two periods between January 1999 and March 1999. The average (of three measurements) of aerosol size-resolved chemical composition data, obtained during the second period is used for the present study. During the second period, the average ambient temperature and humidity were 30 ˚C and 42%, respectively. For the present calculations, the aerosols are assumed dry for this humidity level. The total aerosol mass size distributions have been described using bimodal log normal distribution with a fine mode and a coarse mode. Coarse mode was found to contribute approximately 69% of the total PM_{10}. For the calculations, fine mode MMD has been assumed as 0.7 μm with GSD 1.4. Coarse mode MMD was approximated as 5 μm with GSD 1.5 and mass fraction of 0.69. The aerosol size distributions described above were assumed to correspond to dry particles, with specific soluble contents in fine and coarse modes. Soluble fractions in fine and coarse mode were assumed as $(NH_4)_2(SO_4)$, NaCl, $NaNO_3$, NH4NO_3, Ca $(NO_3)_2$, and soluble complexes of K, Ca, and Mg. Non-sea salt sulphate is assumed as $(NH_4)_2(SO_4)$, sea salt sulphate is assumed as Na_2SO_4, Cl is assumed as NaCl, the remaining Na is assumed as $NaNO_3$, the rest of the fine mode NO_3 is assumed as NH_4NO_3, and the coarse mode NO_3 is assumed as $Ca(NO_3)_2$.

Since, there is no information regarding remaining portions of the Ca, Mg, K and associated compounds, these are assumed to be instantly dissociated in the water. For other parameters (density, molecular weight, etc.), they are assumed as carbonates for the present calculations. The interactions between the compounds or compounding effects of mixture of compounds, if any are ignored in the model. The normalized size distributions of the soluble and non-soluble core are depicted in Fig. 8. The detailed information of mass fractions and the size distribution parameters of each compound can be obtained from the reference (Venkataraman *et al.*, 2002). The aerosols were assumed as internally mixed; therefore the effect of external mixing assumption is separately examined.

RESULTS

Single particle growth

Fig. 1 shows the volume growth curve of particles containing water soluble fractions. The particles are initially crystalline, and growth is calculated under typical lung conditions of temperature and relative humidity (Ta = 37˚C, RH = 99.5%). The soluble fraction is assumed as

NaCl. The figure indicates the dominant effect of water uptake on the particle volume. For the particles containing a soluble fraction of 20%, the volume growth factors (for 10 seconds growth time) have ranged between 3.82 and 30.12, which corresponds to the diameter change in the range of 1.56 to 3.11. Particles with size 0.1 μm reach the equilibrium size within 1 sec of the growth. But the particles larger than 0.1 μm are not attaining the equilibrium size even after 10 sec. Normal human breathing periods of rest are about 4-5 sec (15 breaths per min). This shows that the bigger particle does not attain equilibrium growth in a single breath. Computational studies on droplet growth have shown a similar pattern in the growth rate. (Seinfeld and Pandis, 1998). But in actual conditions, the particle that is not deposited may not exhale in one breath. This may occur when particles carried by fresh air mix with air residing in the respiratory airways (Sarangapani and Wexler, 2000). But the ultrafine particles (< 100 nm) will equilibrate with the water vapor inside the lung within 1 sec and grow to the maximum size in a single breath. As the RH increases, the forcing term in the Eqn. (2) will be greater and consequently induces the larger growth rate. But, the growth rate is limited by transport of vapor, as well as heat [Eqn(3)]. Therefore, particles may not grow faster at higher RH. However, the RH of the medium will decide the equilibrium size of the particle. Since the residence time of the upper respiratory tract at normal breathing conditions is well below 0.5 sec (see Fig. 2), the above fact also indicates that the variations in RH profiles in the upper part of the respiratory tract may not have much effect on bigger particles (> 1.0 μm). The variations in the RH profiles may not have much effect on initial growth phase, which will be limited by the kinetic factors of Eqns. (2) and (3). But since, the smaller particle reaches equilibrium much below or around 0.5 sec, the relative humidity variations in the upper respiratory tract have an effect on particle size (Broday and Georgopolous, 2001).

Fig. 1 also shows that bigger particle (> 1.0 μm) growth is slow to have any impact on the upper regions of the respiratory tract, especially for the extra thoracic region. This is because the cumulative residence time in the upper lung regions are below 0.1 sec. However, coarse-particle water uptake and growth have impacts on the lower respiratory tract due to the large cumulative residence time. Fig. 1 compares the growth of particles having different fractions of soluble material. The figure also shows the growth profiles of pure soluble particles and of particles with 20% soluble fraction. Particles with less soluble material have delayed growth rate and relatively small increase in equilibrium size by water uptake. Excluding the organic carbon, the soluble fraction in the typical Indian urban aerosol can vary from 15-30% (Venkataraman *et al.*, 2002). This soluble fraction may significantly affect the particle respiratory deposition due the water uptake. The hygroscopic growth of ambient aerosols and modification of respiratory deposition is investigated in detail in the following sections.

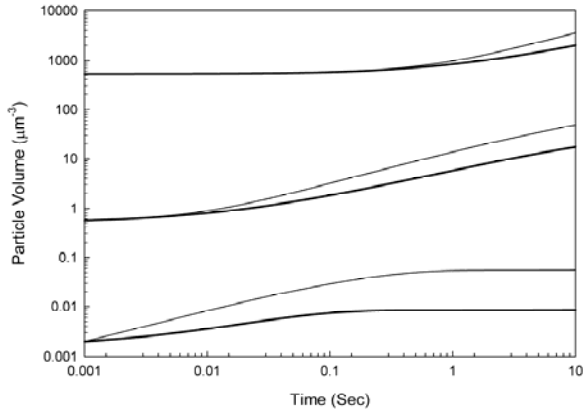

Fig. 1. Volume growth curves for particles of different initial diameters with soluble fractions (a) 0.2- thick line, and (b) 1.0- thin line. The top two curves are for particles with initial dry diameter of 10 μm, middle two curves with a diameter 1 μm, and bottom curves with initial diameter 0.1 μm.

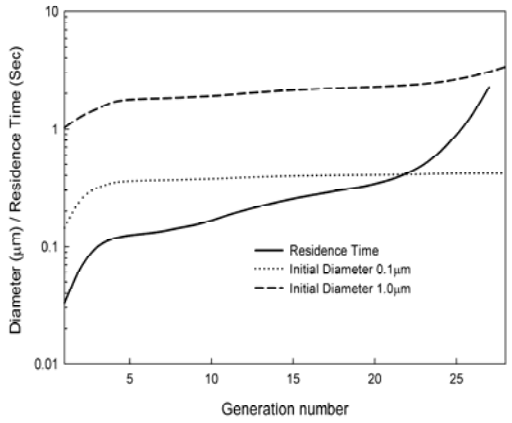

Fig. 2. Residence time and growth rate of particles with soluble material in respiratory tract. Generation number #1 is the equivalent of extra-thoracic, and #3 is trachea. Humidity is 99.5%. Respiratory conditions are 820 cm^3 tidal volume and breathing frequency of 22 per min.

Fig. 2 depicts the particle growth in the respiratory tract during inhalation with the respiratory conditions described above. The growth curves reaffirm the earlier discussion about course and the ultrafine particle growth in the respiratory tract. While particles 0.1 μm in size reached the equilibrium size in the first four generations of the respiratory tract; they did not reach a stable diameter even after the completion of the inhalation part of the breath cycle. The growth rate of particles per generation for the coarse particle is found to be two maximums at the generations between 1 to 4 and 22 to 27. This is proportional to the residence time available for the particle growth in each generation. For the respiratory parameters considered, the maximum residence

time is observed at the generations 1 to 4 and in 20 to 27. This pattern of growth will effect the particle deposition in the lower respiratory tract, rather than in the upper airways.

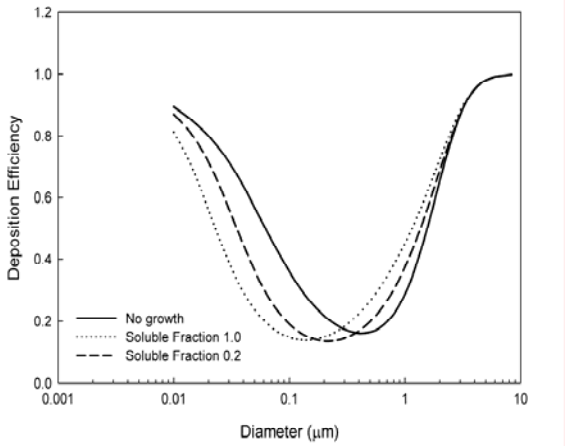

Fig. 3. Effect of water soluble fraction on total deposition efficiency of particles in the respiratory tract. Two fractions of soluble matter are considered 1 and 0.2. Respiratory conditions are 820 cm^3, tidal volume and breathing frequency of 22 per min.

Deposition efficiency and particle growth

The role that water uptake and growth plays on the deposition of inhaled particles as they travel through the respiratory region is now examined. The deposition efficiencies at various conditions can be computed using the model described in the previous sections. The breathing condition simulated here corresponds to a tidal volume of 820 cm^3 and a respiration frequency of 22 per min. The deposition efficiencies incorporating the results for either, considering hygroscopic growth, or ignoring it, is shown in Fig. 3. To neglect the effects of growth, the particle diameter and density were kept constant throughout the simulation to incorporate hygroscopic effects; those parameters were computed as a function of residence time in each generation using Eqns. (2) and (3). The total deposition curve for the dry particle is left-shifted and widened when the hygroscopic growth is included. The left shift is because as the particles grow, the smaller diameter particles behave as the bigger diameter particles, and accordingly, the deposition efficiency of the particles change. The deviation from the no-growth curve is not symmetrical; the curve shows maximum deviation is observed for the particle diameter between 0.1 and 0.01 μm, and lesser at the coarser particle. As discussed above, the coarser particles grow slowly and they are effectively removed by the upper part of the respiratory tract. But the ultrafine particle (diameter < 0.1 μm) grows rapidly and has maximum water uptake and growth effect. Since deposition mechanisms for the bigger particles are impaction and sedimentation, the

size growth may be partly offset by the reduction in density of the particle. The deposition efficiency is increased for particle diameter greater than 0.2 μm and reduced for smaller particles. This is mainly due to the difference in mechanisms involved in deposition of smaller and coarse particles. The increase in particle size can reduce the deposition due to diffusion mechanism, and can increase deposition efficiency by both sedimentation and impaction. The curve for the 0.2 μm fraction of soluble material shows significant deviation from the dry-particle curve. This points out that the hygroscopic growth effect has to be taken into account for accurate prediction of deposition of ambient aerosols.

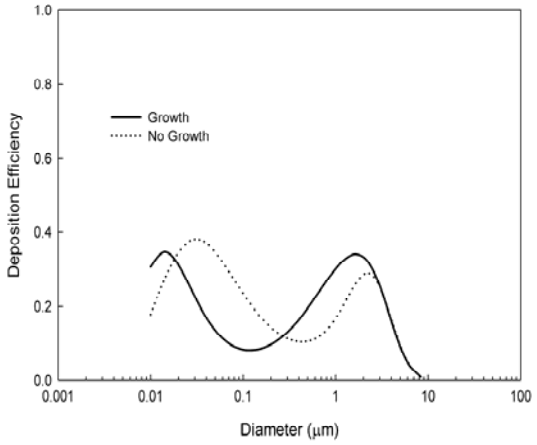

Fig. 4. Effect of water soluble fraction on alveoli deposition efficiency of particles. The soluble matter is assumed as NaCl with a fraction 1. Respiratory conditions are 820 cm^3 tidal volume and breathing frequency of 22 per min.

The regional deposition efficiency curves (Figs. 4 and 5) show the particle deposition in the alveoli and the trachea bronchial (TB) region of the respiratory tract. Similar to the total deposition, both the curves are shifted to the left. But the deviation from the dry particle curve from the coarse particle curve is more for alveoli than for TB. This is because of the increased residence time in the alveoli region discussed above. However, while there is no increase in particle deposition for particles greater than 2 μm for alveoli, there is moderate increase in the particle deposition in TB region. This indicates that it may be possible to use the particles bigger than 2 μm with hygroscopic content to increase the target deposit in TB region.

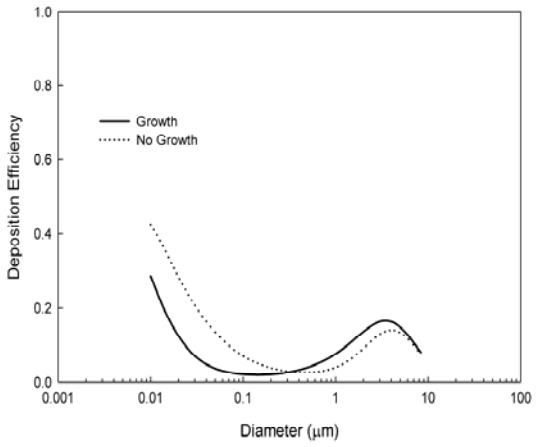

Fig. 5. Effect of water soluble fraction on Trachea Bronchial deposition efficiency of particle. The soluble matter is assumed as NaCl with a fraction 1. Respiratory conditions are 820 cm^3 tidal volume and breathing frequency of 22 per min.

Comparison with experimental observations of deposition efficiency of hygroscopic particles

Comparison between present predictions and experimental results on deposition of hygroscopic particles are given in Figs. 6 and 7. For this comparison, the experimental data of Londhal *et al*. (2006), Tu and Knutson (1984), and Blanchard and Willeke (1984) have been used. Fig. 6 shows the experimental results from Londhal et al. (2006), and Tu and Knutson (1984) along with the model predictions of total deposition efficiencies for insoluble particles in the size range of 0.01 to 1 μm. Deposition computed with the empirical ICRP model (ICRP, 1994) is also shown in the figure. The data shown in Fig. 6 refer to a tidal volume of 1000 cm^3 and 4 s respiration period. The insoluble aerosols tested were DEHS (Londhal *et al.*, 2006), kerosene heater, and aluminosilicate (Tu and Knutson, 1984) particles. The model reproduces satisfactorily the experimental data on insoluble particles over the size range, considering the uncertainties in the measurement. The ICRP model results are also comparable with the present model.

The above lung deposition experiments were also repeated using sodium chloride, which is known to be hygroscopic, and particles are expected to grow in the respiratory airways. Fig. 7 shows the results of the experiments with sodium chloride particles along with model calculations. As discussed in the earlier paragraphs, the sodium chloride curve is shifted to left in comparison with the insoluble deposition efficiency curve. The experimental data are more scattered in the case of NaCl particle than in the case of insoluble particles. In earlier discussion, it is pointed out that for small particles, the changes in humidity and temperature profiles of the respiratory tract have the maximum effect on growth and deposition. Therefore, the inter-subject and inter-experimental variability of the deposition may have enhanced with hygroscopic particles. The

model calculations are within the uncertainty range of the experimental data and the model reproduces the deposition efficiency relationship of the NaCl particle with particle size fairly well. However, the rising limb (0.1 to 1.0 μm) of the model-calculated deposition efficiency curve is found to have deviated more from the experimental data. The calculations with constant density for the particle (See Eqn. (4)) reduced the deviation. The earlier comparisons (Mitsakou *et al.*, 2005) using models having constant density during the growth have also reproduced the experimental data in this size range. Therefore, further investigation is required on the deposition efficiency of hygroscopic particles in this size range.

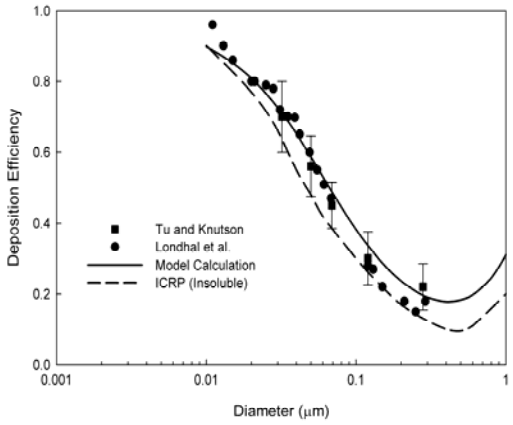

Fig. 6. Total deposition efficiency of insoluble particle in the respiratory tract experimentally determined and calculated. The deposition efficiency is calculated for a tidal volume equal to 1000 cm^3 and a respiration period equal to 4 s.

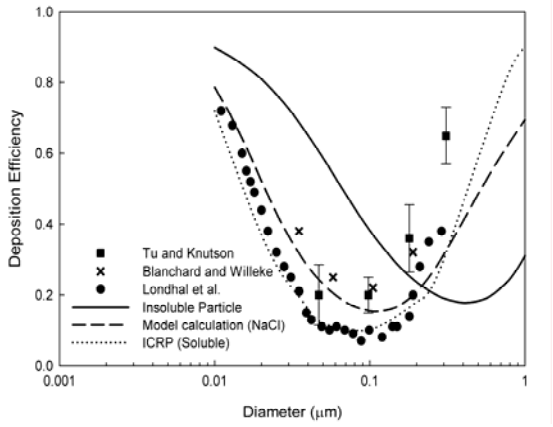

Fig. 7. Effect of water soluble fraction on total deposition efficiency of particle in the respiratory tract, experimentally determined and calculated. Soluble particles are considered as NaCl. The deposition efficiency is calculated for a tidal volume equal to 1000 cm^3 and a respiration period equal to 4.

Also, in this size range (> 0.1 μm), the experimental data deviate considerably from the calculated NaCl efficiency curve. Magnitude of this deviation is greater than that between experimental and model calculated insoluble particle deposition efficiencies. Therefore, the model may produce conservative estimates of the effect of the soluble particles for particles greater than 0.1 μm. Since the difference is moderate and on the conservative side, the model is used further to obtain the lung dose of the atmospheric particles containing soluble substances.

Effect of water soluble compounds on daily lung dose from particulates exposure

The size-resolved deposition efficiency of particulates are integrated over particle size distributions (Fig. 1) to obtain the fraction deposited in each airway generation of the lung. Three respiratory physiological and functional conditions were used for the estimation of the lung dose. In Case 1, respiratory functions were selected to reflect the respiratory physiology of an Indian woman involved in household activities (Varghese *et al.*, 2005). This corresponds to a tidal volume of 820 cm^3 and a breathing frequency of 22 per min. Case 2 represents similar physiological conditions, except for a low breathing rate of 15 per minute. Case 3 represents a healthy male with a functional residual capacity of 3300 cm^3, tidal volume 1500 cm^3 and breathing rate 15 per minute. The calculations were carried out with and without hygroscopic growth.

For Case 1, the total daily mass dose of the exposure to 162 $\mu g/m^3$ for the breathing cycle (tidal volume of 820 cm^3 and respiration frequency 22 per min) for an exposure of 8 hours per day are 113.46, 79.2 and 408.2 μg/day in NP, TB, and alveoli regions of the respiratory system. About 43% of the inhaled particulate mass is deposited in the respiratory tract with 29% in the NP, 6% in the TB, and 8% in the alveoli region. The major part of the particulate mass was in the coarse fraction, where the deposition mechanism of the NP was predominant, and resulted in the peak deposition in the NP region. The total and regional deposition of the particulate matter is given in the Table 1. Comparison of the regional deposition of cases 1 and 2 reveals the influence of the breathing pattern on the lung deposition. The deposition fraction in the alveoli is comparatively high for Case 2 and show a lower NP deposition. For the mass size distribution considered here, the major deposition mechanism is impaction and is strongly related to the flow rate though the NP region [Eqn. (5)]. Larger flow rate due to high breathing rate for Case 2 resulted in higher deposition in the NP region compared with Case 1.

The daily lung dose for particulate matter, considering the growth of the particles is also shown in Table 1. The parameters used for the calculations are similar to the above dose calculation. In all the cases, the total deposition was moderately increased due to the particle growth. But, the regional lung dose was significantly affected by the hygroscopic growth. The dose fractions increased in the alveoli and TB regions. The change in dose was negligible for the

NP region. Since the major portion of particles was coarser (> 0.5 μm), the particle growth was slow to have any impact on the upper regions of the respiratory tract, especially for the extra thoracic region. The slight decrease in the NP region can be attributed to the change in density of the particle, which is a predominant factor in the deposition in the NP region. The increment in the total deposition due to hygroscopic growth is more than the increase in the soluble fraction. This indicates that when the aerosol mixing is internal, the soluble fraction enhances the deposition of the insoluble fraction.

As discussed in the previous section, the coarser particles grow slowly and they are effectively removed by the upper part of respiratory tract; the magnitude of the hygroscopic effect is low for coarse particles compared to the fine particles. Considering the fact that the bigger particle (1 μm) predominates the mass size distribution considered in the present study, and the model is conservative in estimation, these results point out that hygroscopic growth has significant effect on the deposition of ambient aerosols. The results also show that, compared to the alveolar dose, the increase in the deposition of particle growth is high in the TB region. This observation matches with the earlier discussion on the TB deposition that there is no increase in particle deposition for particles greater than 2 μm in size for alveoli, but there is moderate increase in the particle deposition in TB region.

Table 1. Daily respiratory dose of total and soluble fractions of ambient aerosol concentration measured at Mumbai, for various respiratory physiological conditions.

			Total and Regional daily dose μg/day			
			Alveoli	Trachea Bronchial	Nasopharyngeal	Total
Case 1	Total	No growth	113.46	79.22	408.31	600.97
		Growth	142.54	109.63	402.68	654.86
	Soluble	No growth	39.91	21.16	90.94	152
		Growth	52.73	32.9	88.7	174.34
Case 2	Total	No growth	104.67	67.42	236.34	408.44
		Growth	120.91	101.3	237.69	459.9
	Soluble	No growth	36.14	17.2	50.2	103.54
		Growth	43.47	29.82	50.73	124.02
Case 3	Total	No growth	167.85	77.07	540.78	785.7
		Growth	217.47	117.38	538.99	873.84
	Soluble	No growth	60.23	21.04	122.7	203.97
		Growth	79.99	36.67	121.95	238.6
Case 1 (external)	Total	Growth	129.04	95.67	404.73	629.44
	Soluble	Growth	54.56	37.91	90.55	183.02

Particulate mixing was assumed as internal for all dose estimations. Such complete mixing is ideal and does not exist in nature. The lung dose fractions are also estimated using the external assumptions. For this calculation, chemical species are assumed not to be mixed with any other compounds. Change in the diameter and density of the pure compounds are calculated using the growth equations. The results (Table 1) show that, in the case of pure external mixing, the total respiratory deposition is decreased. The total lung dose obtained based on external assumption for Case 1 is found to have deviated 3.8% from the estimates based on internal assumption. But the regional lung deposition varied in the range of 1-12%. However, the deposition of the soluble fraction is increased in the case of pure external mixture. This is due to the increase in growth of pure soluble fractions. But external mixing does not change the deposition pattern of the insoluble core. These results imply that the particle mixing assumption has effects on the estimation of the regional lung dose for the aerosol size distribution considered here.

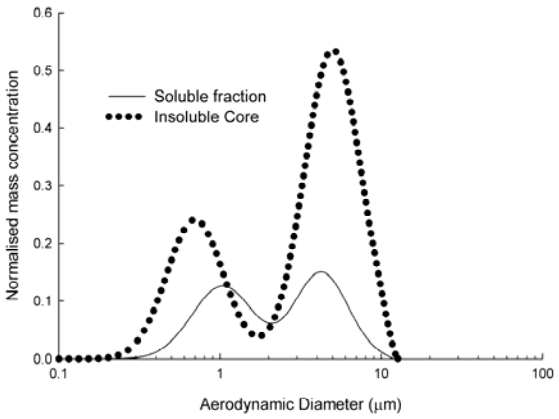

Fig. 8. Normalized mass size distribution of soluble and insoluble core fractions of ambient aerosols measured at Mumbai (Venkataraman *et al.*, 2002).

CONCLUSION

The effect of water-soluble fractions on the particle deposition efficiency in the human respiratory tract is studied with a numerical model. The simulations indicate that the particle can grow up to two times or more of its original size due to water uptake, depending on the quantity of the soluble matter it carries. Water uptake and growth have significant effect on particle deposition efficiency. The ultrafine (< 0.1 μm) range of particles will reach their maximum growth by the time they exit the NP region, whereas larger (> 1.0 μm) particles may still be growing even after the inhalation process is completed. Therefore, soluble ultrafine particle growth has larger effect on the deposition efficiency. The water uptake of particles can change

particle diameter as well as the density, which will affect their deposition.

The model results are compared with experimental results of sodium chloride particles. The model reproduces the major features of the experimental data. The present study also investigated the lung dose of soluble compounds based on the composition of the ambient particulate matter measured in Mumbai, India (Venkataraman *et al.*, 2002). The results indicate that water uptake in the respiratory tract may significantly affect the respiratory dose of the ambient particulate matter containing soluble material. The effect of particle mixing assumptions (external or internal) on the estimation of the respiratory dose of particles containing soluble material is also investigated. It is found that the particle mixing has moderate effect on the estimation of the lung dose for the aerosol size distribution considered.

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