



Particle Size Distribution of Mainstream and Exhaled Cigarette Smoke and Predictive Deposition in Human Respiratory Tract

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ABSTRACT

The particle size distribution of cigarette smoke is an important factor in predicting the deposition fraction of the inhaled particles in various regions of the respiratory tract. Mainstream cigarette smoke is a direct concern for smokers, while the exhaled and side stream smoke contribute to passive (second hand) smoking. The particle size distribution of tobacco smoke for mainstream and exhaled smoke has been studied in this work. This study investigates how smoking behavior, including puff volume and number of puffs, affect the particle size distribution of mainstream cigarette smoke. Scanning Mobility Particle Sizer (SMPS) was used to measure the particle size distribution of mainstream and exhaled cigarette smoke. The values of count median diameter (CMD) mobility size for the first four consecutive puffs were found to be 193 nm, 198 nm, 194 nm and 186 nm. As the volume of a puff increases from 35 mL to 85 mL, the CMD shifted slightly towards the higher particle size ranges, because of increased probability of coagulation and other combination processes. For the exhaled cigarette smoke, the growth factor was found to be 1.5 ± 0.3 with respect to mainstream cigarette smoke. The experimental results of particle size distributions were used in a Multiple Path Particle Dosimetry (MPPD) model to predict deposition patterns in the human respiratory tract. The MPPD model results show the deposition fractions for mainstream cigarette smoke were 0.163, 0.152 and 0.298 for the head, trachea and bronchi (TB) and pulmonary region, and those for exhaled cigarette smoke were 0.273, 0.064 and 0.134 respectively. The total calculated deposition fraction for mainstream and exhaled cigarette smoke was found to be 0.613 and 0.471, respectively.

Keywords: Particle size distribution; Mainstream cigarette smoke; Exhaled cigarette smoke; SMPS; Deterministic dosimetry model; Respiratory tract deposition.

INTRODUCTION

Tobacco smoke is a complex mixture of gaseous compounds and particulates. Current literature shows 4800 identified gaseous and particulate bound compounds in cigarette smoke (Baker, 1999; Perfetti and Rodgman, 2008).

Airborne particulate matter (PM), and especially fine particles, has been associated with various adverse health effects. Environment tobacco smoke (ETS) is an important source of anthropogenic pollution in indoor environments. Cigarette smoke made of gaseous pollutants as carbon monoxide (CO), sulphur dioxide (SO₂), nitric oxide (NO), nitrogen dioxide (NO₂), methane (CH₄), non-methane hydrocarbon (NMHC), carbonyls and volatile organic compounds (VOCs); and particulate matter (PM) (Diapouli *et al.*, 2011; Wang *et al.*, 2012).

In assessing the effects of cigarette smoke on smokers and

nonsmokers, the particle size distribution of the aerosol is an important factor in predicting the deposition fraction of the inhaled particles in various regions of the respiratory tract (Chang *et al.*, 1985). During cigarette smoking there are two dominant pathways of smoke generation. The mainstream smoke which goes as a puff in smoker's mouth through filter and side stream smoke which is emitted in air from the burning end of cigarette (not through filter). Deposition of mainstream cigarette smoke (MCS) in the respiratory tract is a key mechanism for exposure of smokers to smoke constituents and in understanding smoking-related disease (Feng *et al.*, 2007; Moldoveanu and St. Charles, 2007). The particles in Environmental Tobacco Smoke (ETS) are derived from particles in fresh mainstream and side stream smoke. However they undergo mixing and dilution (i.e., dispersion), and follow different routes of exposure over varying time scales in a particular indoor setting. While undergoing dispersion, the median tobacco smoke particle size can shrink as particle mass evaporates or can grow as particles coagulate (Klepeis *et al.*, 2003). The mainstream smoke is a direct concern of smoker, at the same time the exhaled and side stream smoke combined, known as Environmental Tobacco Smoke (ETS) is responsible for

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passive smoking.

Smoking can be simulated as a two-step process. The first step involves puffing of the cigarette and drawing smoke into the mouth where it is held for a finite period of time, as the palate generally closes off the mouth from the rest of the airways during puffing. After this step the second step is inhalation of the smoke into the lungs. Therefore the puffing process can be decoupled from the inhalation process. Various mechanisms have been put forward to explain the deposition pattern of cigarette smoke in respiratory tract, including coagulation, hygroscopic growth, condensation and evaporation, changes in composition, or changes in inhalation behavior (McGrath *et al.*, 2009). The disparity which exists between experimental and predicted data from deposition models has still not been fully addressed in the literature (ICRP, 1994). This may be due to incomplete incorporation of parameters in predictive models for cigarette smoke deposition in respiratory track. The particle concentration in tobacco smoke is generally very high (10^{12} particles per cigarette). Also, the hygroscopic nature of the smoke droplets results in rapid changes in particle diameter via coagulation and condensation (McRae, 1990). The three main mechanisms that affect the behavior of cigarette smoke particles in the respiratory tract are gravitational sedimentation, inertial impaction and Brownian motion (diffusion). Sedimentation and impaction are 'aerodynamic' effects that are important above about $1\ \mu\text{m}$ and increase with increasing size. Aerodynamic effects are negligible for very small particles and thermodynamic effects are negligible for large particles. The deposition of smoke particle depends on the size of particle which may vary due to the high relative humidity condition in the respiratory tract. Despite the small diameter of the smoke particles, smoke deposition efficiencies of 60–80% in the lung have been reported (Baker and Dixon, 2006). In this study an attempt has been made (1) to quantify particle size distributions for mainstream as well as exhaled cigarette smoke; (2) to study the effect of puffing conditions on the particle size distribution of fresh mainstream cigarette smoke (MCS); and (3) to evaluate the deposition of number concentration fraction in various compartments of the human respiratory tract for both main stream and exhaled cigarette smoke.

MATERIALS AND METHODS

Experimental Designing

The present study of cigarette smoke (mainstream and exhaled) analysis is carried out on the popular and widely consumed brands in India. The tar content of the cigarettes used in this study was in the range of 11 to 17 mg per cigarette. The tar deliveries are pack tar values which are generated using ISO puffing conditions. Cigarettes used for these experiments were conditioned in a humidified chamber at $65\% \pm 5\%$ relative humidity (RH) at room temperature ($24^\circ\text{C} \pm 3^\circ\text{C}$) for at least 24 h prior to smoking. The cigarettes were smoked under the ambient laboratory conditions ($45\% \pm 5\%$ RH, $24^\circ\text{C} \pm 3^\circ\text{C}$). The mainstream smoke puffing setup was planned in such a way that it can provide the required smoking parameter i.e., puff volume, puffing duration, and

number of puffs. In the first set of experiments the puff volume was fixed at 35 mL with a flow rate of 1.1 L/min and puff duration was kept for around 2 seconds to know number concentration size distribution on puff by puff basis. The second set experiment was designed to reveal changes in particle size distribution with variation in puff volume. The puff volume was varied from 35 mL to 85 mL by changing the flow rate. Thirty male volunteers in the age group of 25–40 were selected for the experiment. The ethical approval signed by all the volunteers was taken prior to performing the experiment, and all volunteers were smokers. The particle size distribution of exhaled cigarette smoke was measured directly by collecting the smoke in dilution chamber.

Instrumentation

In this study, the GRIMM (Germany) made SMPS-C (Sequenzial Mobility Particle Size analyser) Series 5.400 was used to measure particle size distributions. The cigarette smoke sample passes through a pre-impactor (flow rate of 0.3 L/min) to define the upper cut-off diameter of the particles, and a radioactive charger (Am-241) to charge the particles to a defined charge distribution. Only particles with an appropriate charge and size travel to the sample air outlet, entering the Condensation Particle Counter as a mono-disperse aerosol. As an aerosol first enters the CPC; it is saturated with alcohol vapor as it passes over a heated pool of alcohol (butanol). The vapor-saturated aerosol then flows into a cold condenser, where it is cooled by thermal diffusion. The alcohol condenses onto the particles and the particles grow into droplets large enough to be counted optically. Once the particles have grown to an optically detectable size (typically, 2 to 3 micrometers), they pass through a light beam and scatter light onto a photo detector. The instrument measures particle concentrations in a size of 10 nm to 1083.3 nm in 46 channels. In present study, the spectrometer was operated in size distribution mode to produce number concentration versus size channel plot. The measured real-time number concentration data are transferred at 5 minute intervals to a data storage card (Baxla *et al.*, 2009).

Experimental Setup

The experimental simulation setup under puffing conditions is described in Fig. 1. The mainstream cigarette smoke was passed through a dilution chamber prior to being sampled by the SMPS so that the saturation point of the SMPS was not exceeded. Particulate free nitrogen was used as dilution media and it did not contribute any particulate signature in SMPS. The setup was checked for leaks and the mass flow meter was calibrated using master meter (flow meter) with high accuracy. All flows were controlled with mass flow controllers (Model 2179A; MKS, Andover, MA) with accuracies $> 1\%$ of their operating flow rate. The dilution stage provided a dilution of 140 for the whole system. This dilution ratio provided particle concentrations in the working range of instrument. It also avoids multiple dilution steps and hence measurements were more reliable. Smoke generated under different puff volume conditions were delivered to the dilution chamber and then diluted with particulate free

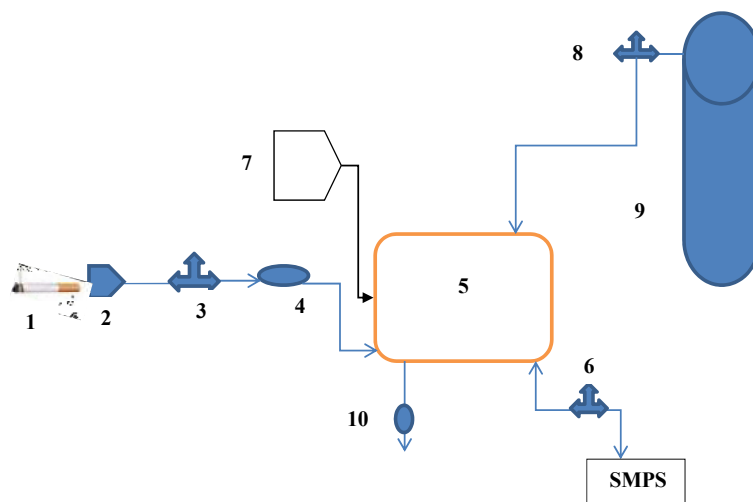


Fig. 1. Mainstream cigarette smoke analysis setup 1) cigarette 2) cigarette holder 3) MCS mass flow controller 4) suction pump (1–3 L/min) 5) dilution chamber 6) flow controller for scanning mobility particle sizer (SMPS) 7) Mouth piece for collection of exhaled cigarette smoke 8) N₂ nitrogen cylinder flow controller 9) N₂ cylinder for dilution 10) pump for dilution chamber evacuation.

air. The dilution chamber was flushed with particulate free air (using HEPA filters) between the puffs measurements to ensure that no particulates were present in the dilution chamber before the subsequent puff was taken.

Respiratory Tract Deposition Model

The Multiple-Path Particle Dosimetry (MPPD V2.11) model developed jointly by the Hamner Institutes for Health Sciences and the Dutch National Institute for Public Health and the Environment has been used for prediction of aerosol deposition fractions in different compartments of the human respiratory tract. During smoking, the cigarette puff is mixed with a volume of ambient air equal to the average lung tidal volume (500 mL) that delivers the smoke to the lungs (Ingebrethsen *et al.*, 2011). This volume mixes with and dilutes the puff smoke as it travels into the respiratory tract. There may be no, complete, or partial mixing of the puff with the dilution air. If x denotes the fraction of the puff volume that mixes with the dilution air, the deposition fraction of the inhaled puff (DF) is found by tracking the MCS particles in the puff and dilution volumes and calculating their losses.

$$DF = DF_D + (1-x) \frac{V_t}{V_p} (DF_t - DF_D) \quad (1)$$

where V_p and V_t are the puff and tidal volumes, respectively, DF_t is the deposition fraction when the dilution air and smoke from the cigarette puff are completely mixed, and DF_D is deposition fraction when the dilution air is filled with smoke particles but the puff volume is void of any particles (i.e., no mixing). The existing version of MPPD allows particle loss calculations for DF_t and DF_D by invoking oral only and oral plus tracheal breathing route options respectively. The modified version of MPPD automatically calculated these deposition fractions to find the deposition fraction of fully mixed cigarette smoke in the lung (Kane *et*

al., 2010).

MCS contains a significant amount of semi volatile components that are distributed between the particulate and vapor phases (Kane *et al.*, 2010). As the smoke puff mixes with air during smoking, the vapor phase becomes unsaturated, causing evaporation from the particle to establish the particle-vapor equilibrium. MCS also contains many hygroscopic components which may cause inhaled smoke particles to grow in size when they enter the humid environment of the respiratory tract. This growth may directly impact the site of deposition in the respiratory tract and the overall deposition in the airways. The model for hygroscopic growth was used in MPPD to include the effect of hygroscopic growth on deposition (Asgharian, 2004).

RESULTS AND DISCUSSION

Puff Wise Particle Size Distribution for Mainstream Cigarette Smoke

The measurement of the number concentrations of size fractionated aerosols generated from cigarette smoke has been carried out. The result shows very slight variation in particle size distribution patterns with number of puffs. Total number concentration of cigarette smoke per puff was of the order of 10^9 dN/mL. This does not include the water associated with the smoke aerosol, or the volatile organic compounds lost due to evaporation. The particle size distribution of cigarette smoke with puff number is presented in Fig. 2. It was observed that all puffs generated particles were well within the range of SMPS with mobility size ranging from 10 to 1082 nm and showed a lognormal distribution (positively skewed). Again it was found that the cigarette smoke particles above $1.0 \mu\text{m}$ sizes were not contributing significantly to the total number concentration. Considering all the puffs it was found that the cigarette smoke particles mostly distributed to submicron size range i.e., $0.01\text{--}1 \mu\text{m}$ which is in agreement with the earlier

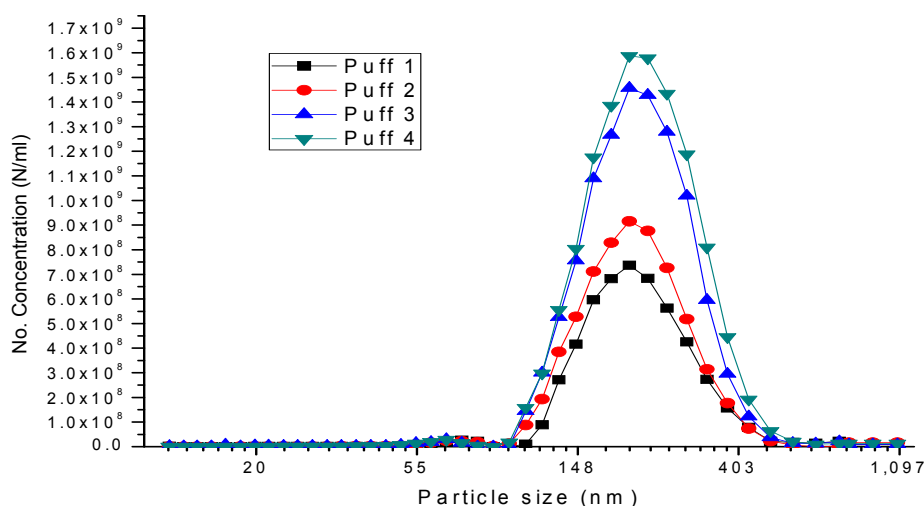


Fig. 2. Mainstream cigarette smoke particle size distribution for initial four puffs, puff duration 2–3 second at flow rate of 1.1 ± 0.01 L/min.

published report (Chang *et al.*, 1985). Number concentration of mainstream cigarette smoke puff number wise was studied for 35 mL puffs which were drawn at a flow rate of 1.1 L/min. The particle size distribution of puff wise mainstream cigarette smoke revealed that there was no significant effect on distributions with puff number. From the particle size distributions, the count median diameter is determined using the cumulative distribution plotted as the percent of the total number concentration versus the stage cutoff point mobility diameter, and using interpolation between the cut point just above and just below the 50% mark to find the median. The total concentration was determined by summing the concentration in each size range. The count median diameters (CMD) for initial four puffs were 193 nm, 198 nm, 194 nm and 186 nm (Fig. 3). No systematic increasing or decreasing trend in CMD values of mainstream cigarette smoke was observed with puff number. The geometric standard deviation (GSD) values for mainstream cigarette smoke were found in range of 1.42 to 1.46. This reveals that the previous puff does not affect significantly the distribution of particle number concentration distribution of the next puff. The physical factor of such distribution may arise due to various physiochemical conditions during puffing behavior. As the number of puffs increases, the residence time for smoke to travel through the cigarette rod decreases which leads to decrease in coagulation rate. Hence number concentration of particles increased with the puff number.

Effect of Puff Volume on the Particle Size Distribution of Mainstream Cigarette Smoke

The variation in particle size distribution with puff volume was investigated as part of this study. In this study we have used four different puff volumes by increasing the flow rate (35 mL, 50 mL, 65 mL and 80 mL) and analyzed the resultant cigarette smoke with an SMPS. It was observed from the results that smaller the volume of smoke puff, more the number of particles accumulated in lower sizes. In other words, as the volume of the puff increases the

number concentration accumulates towards slightly higher size range. The experimental outcome of puff volume versus number size distribution and cumulative distribution are shown in Figs. 4 and 5 respectively. At all puff volumes particle size distribution was uni-modal with positive skewed. The results show that as the volume of the puff increases there is higher concentration of smoke particles in the dilution chamber which enhance the probability of coagulation and other combination processes and thus increase the average particle size. No correlation was found between particle size and puff volume of mainstream cigarette smoke. The slight shift of particle number in higher size range observed may be due to coagulation in dilution chamber.

Particle Size Distribution for Exhaled Cigarette Smoke

Exhaled cigarette smoke is major contributor to environmental tobacco smoke and second hand (passive) smoking. The particle size distribution of exhaled cigarette smoke was investigated using scanning mobility particle sizer (SMPS). Exhaled puffs were passed in the dilution chamber and diluted so that instrument saturation should not occur. The particle size distribution of exhaled smoke and the cumulative distribution plots are shown in Figs. 6 and 7 respectively. The number concentration for exhaled cigarette smoke was found to be in the range of 10^9 numbers per mL. The count median diameter (CMD) for exhaled smoke was found 0.23 ± 0.05 μm range while the geometrical standard deviation (GSD) was found in the range of 1.85 to 2.10. The ratio of mainstream smoke CMD to exhaled cigarette smoke (growth factor) was found to be 1.5 ± 0.3 which may vary from person to person and also depends on puffing and inhalation behavior. Hygroscopic coagulation is the dominant parameter for growth factor. In practice when cigarette smoke is exhaled into the atmosphere (such as an indoor room) it will mix with side stream smoke and be diluted to form environmental tobacco smoke (ETS) which may undergo deposition by normal respiration. In such case it will get diluted according to ventilation and space of the room.

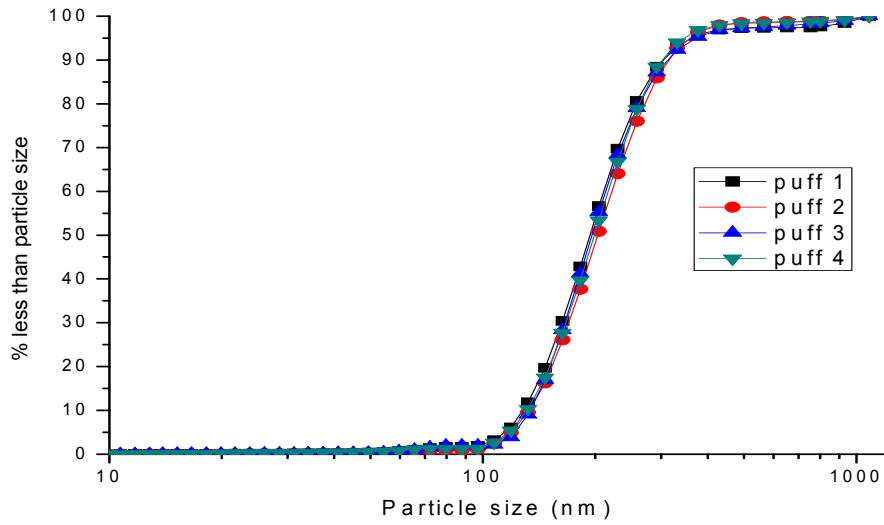


Fig. 3. Mainstream cigarette smoke cumulative size distribution on log probability graph for initial four puffs.

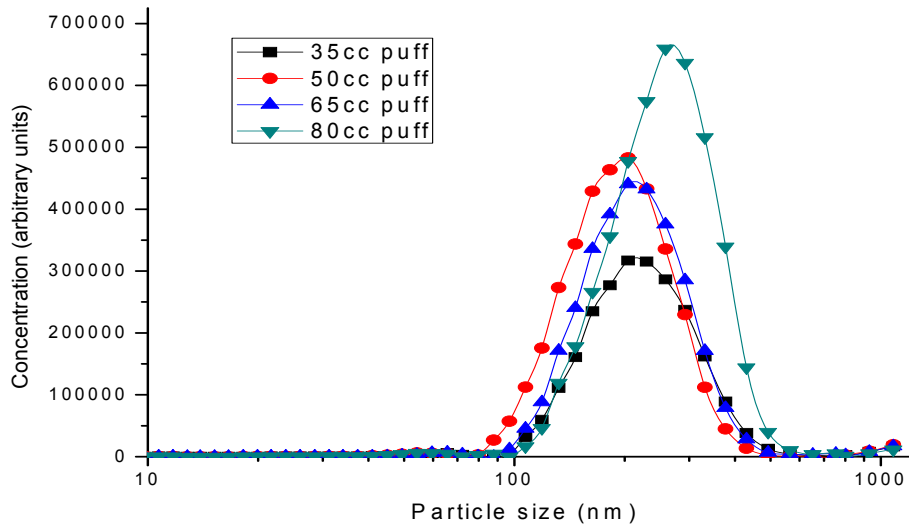


Fig. 4. Particle size distribution for various puff volumes (35 mL, 50 mL, 65 mL and 80 mL).

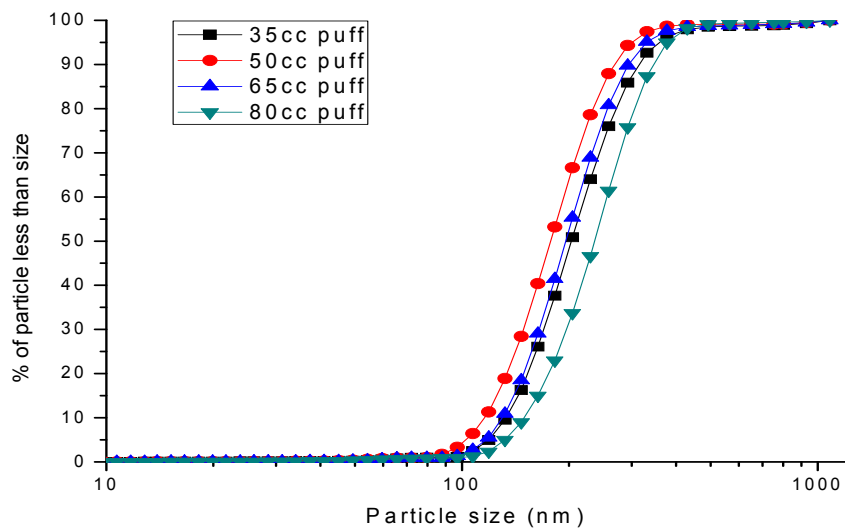


Fig. 5. Cumulative number size distribution of mainstream cigarette smoke for different puff volume (35 mL, 50 mL, 65 mL and 80 mL).

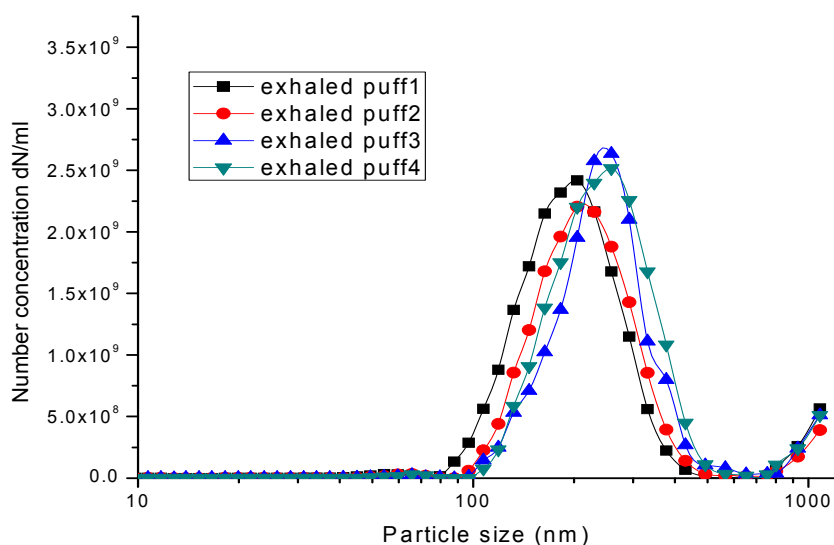


Fig. 6. Particle size distribution of puff wise exhaled cigarette smoke measured with SMPS.

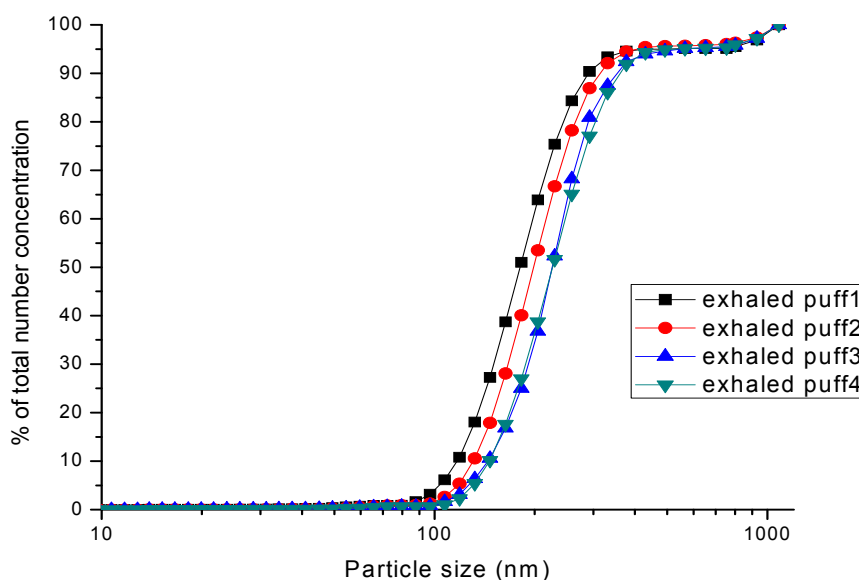


Fig. 7. Cumulative distribution plot of exhaled cigarette smoke for initial four puffs.

Predicted Respiratory Tract Deposition for Mainstream and Exhaled Cigarette Smoke

From the normal smoking behavior it was observed that a smoker takes nine puffs on an average from a cigarette within a time span of three to four minute. In MCS particles contained within a puff are at a very high concentration and undergo rapid coagulation, which leads to a reduction in number concentration and an increase in particle size. Coagulation occurs more rapidly in the oral airways during the puff hold and will decrease on mixing with the dilution air. Additional coagulation occurs during inhalation but particles quickly approach an asymptotic concentration and size, with little or no change during exhalation. Similarly the exhaled cigarette smoke gets diluted in room conditions and may be inhaled again during normal breathing. But there is no data to quantify the extent of dilution and change in particle size distribution with time and space. During normal

respiration the deposition of particle in various respiratory compartments is shown in Fig. 8 (generated using MPPD model), which shows how particle size is responsible for deposition in respiratory track.

Predicted Respiratory Tract Deposition for Mainstream Cigarette Smoke

The Yeh/Schum symmetric lung model (Yeh *et al.*, 1979, Asgharian *et al.*, 2001) was used for the modified MPPD with input parameters as follows: puff volume 35 cm³; concentration of undiluted cigarette smoke 15000 mg/m³; time for exposure to one cigarette for 3 minutes, functional reserve capacity (FRC) of 3300 mL, upper respiratory tract volume of 50 mL, and particle density of 1 g/cm³ (Kane *et al.*, 2010). As accurate data were not available, the default values for total inhalation volume (500 mL) and breathing period of 5 seconds (2 seconds inhalation, 1 seconds pause,

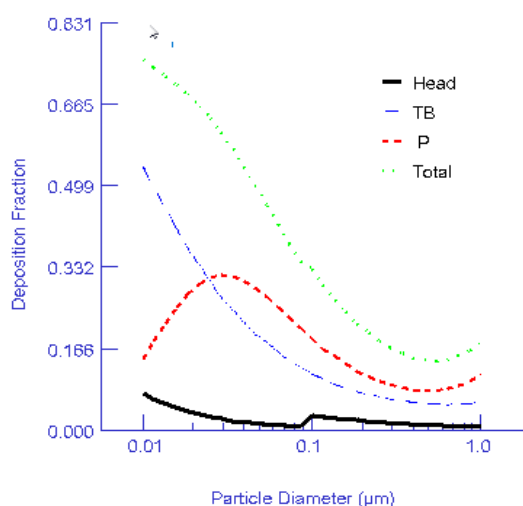


Fig. 8. Predicted deposition fraction for different size particle size using MPPD model.

and 2 seconds exhalations) were used. It was further assumed that 20% of smoke fully mixed with dilution air and there was a 15% deposition fraction in oral cavity. The value of CMD for mainstream smoke was taken 0.185 µm and GSD 1.46. The initial aerosol size distribution used in the calculation was taken from the SMPS results. For these calculations, the geometric standard deviation (GSD) has been calculated using the “probit” method, where the data are plotted as the cut point diameter versus cumulative percent on a log-probability scale and the GSD is given by the slope of the best-fit line through the three highest probability diameters (Hinds, 1999).

The MPPD model predicts how the deposition fraction for the mainstream cigarette varies with particle size in various respiratory compartments i.e., head air ways, trachea and bronchiolar (TB) and pulmonary alveolar are shown in Fig. 9. The total deposition of cigarette smoke in the respiratory system was estimated using the MPPD model. The deposition fractions for mainstream cigarette smoke were 0.163, 0.152 and 0.298 for head, TB and pulmonary

region respectively. Total deposition fraction for entire respiratory tract was 0.613 for the mainstream cigarette smoke. It was also found that the deposition fraction was higher for the size ranges 0.01 to 0.1 micron particle size and the cigarette smoke particle mainly distributed in the size range 0.1 to 1.0 micron. For mainstream cigarette smoke the deposition fraction was highest in the pulmonary region which may be due to higher flow rates, lower size range particle distribution and longer residence time.

Predicted Respiratory Tract Deposition for Exhaled Cigarette Smoke

The input parameters for calculating the deposition of exhaled cigarette smoke in MPPD (V2.11) model were chosen to as close as possible to simulate practical conditions. The Yeh/Schum symmetric lung model was used for the modified MPPD with input parameters as follows; count median diameter CMD 0.25 µm, functional reserve capacity (FRC) of 3300 mL, upper respiratory tract volume of 50 mL, and particle density of 1 g/cm³, breathing route used was nasal. The value of CMD for mainstream smoke was taken 0.25 µm and GSD 2.1. The initial aerosol size distribution used in the calculation was taken from the SMPS results. The MPPD model predicts how the deposition fraction for the exhaled cigarette smoke varies with particle size in various respiratory compartments i.e., head air ways, trachea and bronchiolar (TB) and pulmonary alveolar was depicted in Fig. 10. Total deposition of cigarette smoke in entire lung due to exhaled cigarette smoke obtained by applying MPPD model was found. The deposition fractions for exhaled cigarette smoke were 0.273, 0.064 and 0.134 for head, TB and pulmonary region respectively. Total deposition fraction for entire respiratory tract was 0.471 for the exhaled cigarette smoke.

CONCLUSIONS

Though there are several limitations in this type of study related to experimental method and application of the deterministic dosimetry model still it is reasonable to draw

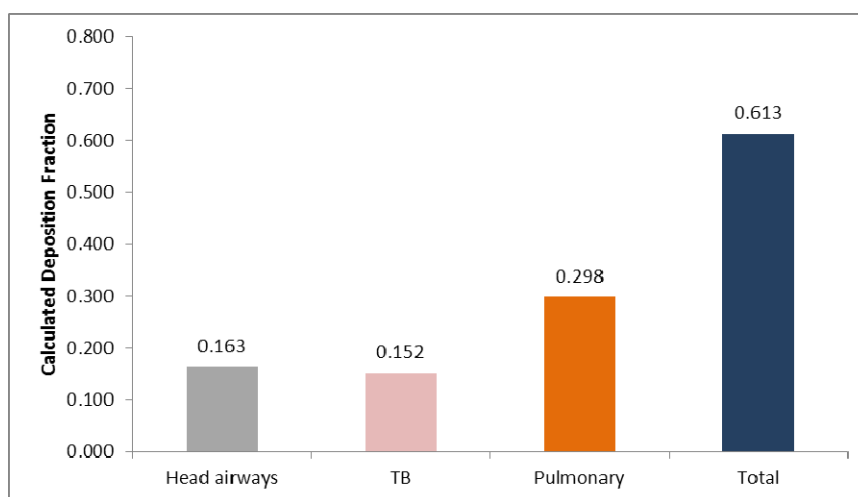


Fig. 9. Calculated deposition fraction for Mainstream cigarette smoke in Entire lung using MPPD model.

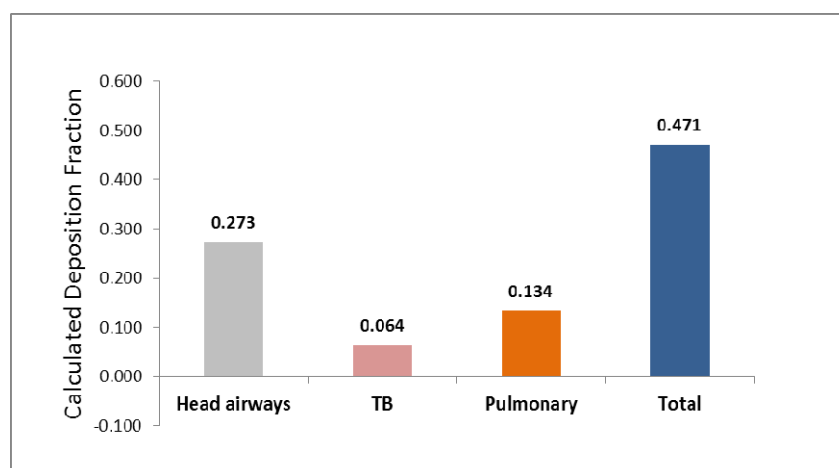


Fig. 10. Deposition fraction for exhaled cigarette smoke for entire lung using MPPD model.

some conclusions from these data. Size fractionated smoke particles emitted by cigarettes have been measured under a variety of conditions. The values of CMD of the mainstream smoke for initial four consecutive puffs were found to be not changing significantly, which indicates that there was no significant effect of increasing puff numbers on particle size distribution. As the puff volume increases more particles stand to be coarser resulting in slight increase in CMD. Exhaled cigarette smoke shows a growth factor of 1.5 ± 0.3 indicating coagulation and hygroscopic growth of smoke particles in the respiratory system. The MPPD model results show highest fraction of mainstream cigarette smoke were deposited in the pulmonary region which may be due to higher flow rate and lower size range particle distribution along with higher residence time in the lower respiratory system. For exhaled smoke the deposition fraction was found to be highest in head airways due to nasal inhalation and larger particle size compared to mainstream cigarette smoke.

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