



Environmental inhalants from tobacco burning: Tar and particulate emissions

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ARTICLE INFO

Article history:

Received 1 June 2018

Revised 21 July 2018

Accepted 31 August 2018

Keywords:

Carcinogenic

Cigarette smoking

Particulate matter

Tar

Tobacco

ABSTRACT

Cigarette smoking is credited for decreasing the world population annually by about 1%. This paper therefore explores the carcinogenic and mutagenic residue (tar), and particulate matter from the thermal degradation of tobacco cigarettes coded, SPM and ES1, at a residence time of 2.0 s at 1 atm. This study was carried out in the temperature range 200–600 °C with nitrogen as the pyrolysis gas. Field emission gun scanning electron microscope was used to image the nature of particulate emissions from tobacco smoke. It was shown that tobacco smoke particulates are ultrafine; ~22 and 28 nm for SPM and ES1 cigarettes respectively. Particle deposition fraction in the human lung and pulmonary lobes was simulated using the Multipath Particle Deposition (MPPD) model. The ultrafine particulates if inhaled are grave precursors for various respiratory health ailments. Maximum tar yield was produced at ~400 °C. Thus, designing cigarettes that may be smoked at temperatures lower than 400 °C may be beneficial to the tobacco smoking community. From MPPD model runs, it was found that the pulmonary tissue retained the highest fraction (0.448) of particles of 22 nm geometric diameter in comparison to 0.418 fraction of the slightly larger particles of 28 nm geometric diameter from ES1 cigarette. This implies that the respiratory system has a poor clearance of particles of smaller geometric diameter. Thus, extremely ultrafine particulates are of grave concern to cigarette smokers.

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Introduction

One of the most controversial poisonous plant, dating back to the 16th century, first introduced to Europe by Christopher Columbus and subsequently to the rest of the world is tobacco [1]. The contemporary cigarette – an industrial form of tobacco, involves more than simply tobacco because of the introduction of additives during cigarette manufacture [1]. The yields of tar in mainstream smoke of commercial cigarette brands are usually measured with smoking machines under highly standardized conditions and must comply with regulatory limits set by tobacco regulatory authorities in various countries [1,2]. It is, however; doubtful if actual representative human smoking behavior can be achieved that may provide accurate simulations of cigarette smoke exposure to humans owing to various factors such as the residence time, puffing

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speed, and the air/liquid interface [2,3], especially in the case of second hand smokers. Environmental cigarette smoke (EST) from a burning cigarette does not necessarily pass through a filter. Therefore, this work is remarkable in establishing the particulate size of smoke particles that might eventually reach second hand cigarette smokers from a burning cigarette. It is worth noting that the pyrolysis of tobacco cigarette is quite similar to the pyrolysis of other forms of biomass components including pesticides; permethrin and captan [4,5]. Nevertheless, this study will restrict itself to material characterization of tobacco smoke.

The focus in this study is primarily on particulate emissions, tar generation and char yields of two commercial cigarettes sold in most parts of the world under conditions representative of cigarette smoking [2,6]. The tobacco thermal char and tar yields are competing processes during tobacco burning [2] and therefore their formation during cigarette smoking is important in predicting temperatures that should be avoided by cigarette manufacturers for the benefit of the cigarette smoking community. This may inform cigarette manufacturers to possibly produce less harmful cigarettes.

Depending on their origin, fine particulates of cigarette smoke may serve as carriers for carcinogens such as benzo[a]pyrene by absorbing them on the surface of particulates [7]. Both long-term and short-term exposure to inhalable particulate matter [8] are linked to injurious effects on human health such as cardiovascular, pulmonary, neurological morbidity and mortality in addition to premature delivery, birth defects and death [9,10]. Although a single cigarette is small in size and typically weighs less than 1 g, it is capable of emitting between 7 and 23 mg of PM_{2.5} when smoked, depending on the smoking conditions and the type of cigarette brand [11]. It is estimated that one cigarette exposes the human respiratory tract to between 10–40 mg particulate matter [8], and have a mean diameter of between 0.1 and 2 µm [12].

The detrimental impact of smoking on public and biological health has been widely documented in literature since the 1960s [13]. In the United States alone, it has been estimated that tobacco smoking causes approximately 400,000 premature deaths per year [14]. In 2015, smoking caused more than one in ten deaths worldwide, killing an estimated 6 million people with a global loss projected at 150 million disability-adjusted life-years [15]. If the current smoking trends is not checked, then tobacco may take out about 1 billion people by the end of the 21st century, with half of these deaths occurring before 70 years of age, especially in developing countries [16]. The major target organ systems in which non-cancer effects of smoking occur include the respiratory system, cardio-vascular system, reproductive system, the eyes, and the nervous system [17,18]. Passive smoking on the other hand is a significant health hazard to children and nonsmoking adults [3] and this is because it is responsible for cases of sudden infant death syndrome, asthma, middle ear infections and meningitis, among other grave diseases such as cardiac arrest, emphysema and cancer of the lungs [19].

Therefore, estimating the fraction of particle deposition in the human lung, following exposure to tobacco particulate matter, is critical towards understanding the risks associated with exposure of airborne pollutants [20]. Diseases initiated by inhalation of various biological particles depend on the nature and size of particles, and also on the number of particles inhaled as well as the site of their deposition in the respiratory scheme [21]. Generally, the disposition of particulates in the body tissues is determined by their rate of diffusion from the tobacco smoke to the tissues, the ability of the tissues to retain the inhalants, and the rate of elimination of the particulates by chemical action, exhalation, metabolism and consequent elimination [22].

This study attempts to derive the relationship between the smoke condensate (tar) and tobacco charcoal (char) at various pyrolysis temperatures that may particularly affect second hand cigarette smokers in general. The temperature at which maximum tar yields are produced is critical in designing cigarettes that can be smoked at lower temperatures and thus minimize the inhalation of tobacco harmful compounds which are usually produced at elevated temperatures as tobacco tar. These harmful compounds contained in tar include carcinogens, mutagens, and particulate emissions which are well-known progenitors for severe biological health diseases afflicting the cigarette smoking community. Additionally, human exposure models are also fundamental in designing exposure estimates for use in epidemiological research because of the high interest in both public and environmental health.

Materials and method

Silica gel (150–200 µm) of ≥99.9% purity used to adsorb tobacco smoke particulates in this study was purchased from Sigma Aldrich Inc. (St. Louis, Missouri, USA). Processed tobacco – commercial tobacco (ES1 and SPM, for confidential reasons cannot be named) of various masses 20 ± 2 , 40 ± 2 and 60 ± 2 mg were separately weight and packed in a quartz reactor of volume ≈ 1.6 cm³ at a residence time of 2.0 s at 1 atm, which is consistent with the ISO smoking regimes - International Organization for Standardization of cigarette smoking [2]. The smoking apparatus employed in this study contains various units: the reactor compartment which houses a muffle furnace and the quartz reactor, and the temperature control console which controls the pyrolysis temperature within a temperature gradient of ± 5 °C.

The respiratory tract deposition fractions in humans was performed using Multipath Particle Deposition (MPPD) Model, version 3.04 [23]. This morphometric model uses statistical relationships of airway parameters to reconstruct the lung geometry [24]. The human model selected for this simulation was Yeh/Schum 5-lobe. The other parameters chosen were; tidal volume 1143 mL and breathing frequency of 17.5 mL/min [22] which is consistent with cigarette smoking conditions. Input data included the size of the particulate (count median diameter - CMD), assuming upright body orientation and constant exposure of tobacco smoke at a given puff. The breathing simulation model selected for this study will be discussed briefly with the aim of supplementing experimental data.

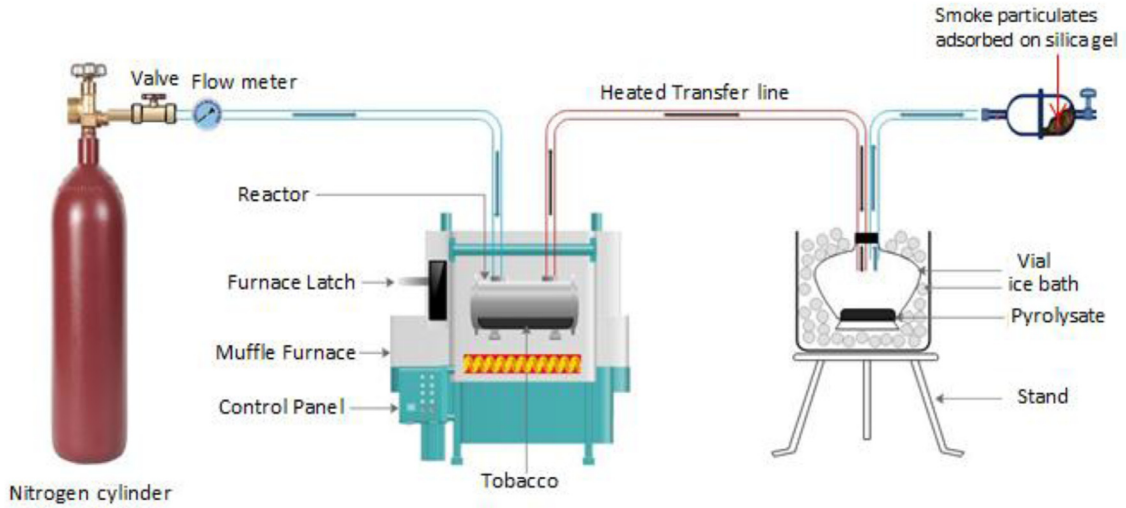


Fig. 1. Reactor assembly and pyrolysate trapping apparatus.

The thermal degradation of tobacco

The pyrolysis temperature was varied at intervals of 50 °C between 200 °C and 600 °C for a total pyrolysis time of 5 minutes [25,26]. The pyrolysis gas was nitrogen. To ensure a near ideal inert pyrolysis environment, the reactor system was flushed with nitrogen gas for 5 minutes to remove any traces of oxygen. In order to investigate the particulate nature of tobacco smoke at 600 °C, 30 ± 2 mg of tobacco was pyrolyzed and tobacco smoke particulates trapped on silica surface as illustrated in the apparatus presented in Fig. 1. The mass of silica used for this study was 50 ± 2 mg. The weight of tobacco smoke particulates adsorbed on silica was determined by the method of difference. On the other hand, tobacco tar was collected via a delivery tube as volatile gases-phase components condensed in an ice bath; modified from Wynder et al. [27] and weighed at the end of each experiment. Six experiments were conducted in replicates to ensure reproducibility and validity of data.

The reactor system

The reactor used is a muffle furnace (Thermo-Scientific Inc., USA) with an internal heating compartment of dimensions of 14 × 13 × 12 cm. The muffle furnace has a temperature regulating knob with a temperature scale ranging from 200 to 1000 °C. The heating compartment, the muffle furnace has an inlet which allows the nitrogen gas tube to pass through into the compartment and an outlet which allows the delivery tube to pass through from the reactor into the collecting vial in the ice bath to trap the tar. The sample holder in the heating compartment is a tubular quartz reactor which can withstand high temperatures of up to 1200 °C. The reactor assembly is presented in Fig. 1.

The residence time was determined from the conventional modified ideal gas formula Eq. (1).

$$t_0 = \left(\frac{\pi r^2 L}{F_0} \right) \left(\frac{T_1}{T_0} \right) x \left[1 + \frac{P_d}{P_0} \right] \quad (1)$$

where t_0 is the residence time, F_0 is the flow rate of the pyrolysis gas and P_d is the pressure difference between the inlet pressure and the pressure inside the reactor. Ideally, the pressure difference is 0 because the ambient pressure and the reactor pressure are supposedly similar (~1 atm) while, L and r represent the temperature, length of the reactor, and the radius of the tubular reactor, respectively. The subscript ₀ denote original parameters (ambient) while the subscript ₁ denotes the parameters inside the reactor.

Scanning electron microscopy analysis

About 5 mg of tobacco particulates adsorbed onto silica gel was introduced into 1 mL methanol and gold grids dipped into the prepared thermal tar sample [28]. Tweezers were used to pick the gold grids from the particulate adsorbed on silica. The sample was then adhered to aluminum SEM stubs with a carbon tape and subsequently gold coated using a Quorum Q150 RES sputter coater. The grids were allowed to dry in air before putting them into the analysis chamber of a Zeiss Ultra Plus (Germany) field emission gun scanning electron microscope (FEG SEM). All images were taken at an angle of 45° to increase the definition of the surface morphology [29]. The images (micrographs) were then saved for further analysis and comparison at various magnifications [30]. Image J software was used to determine the size of the smoke particulates and

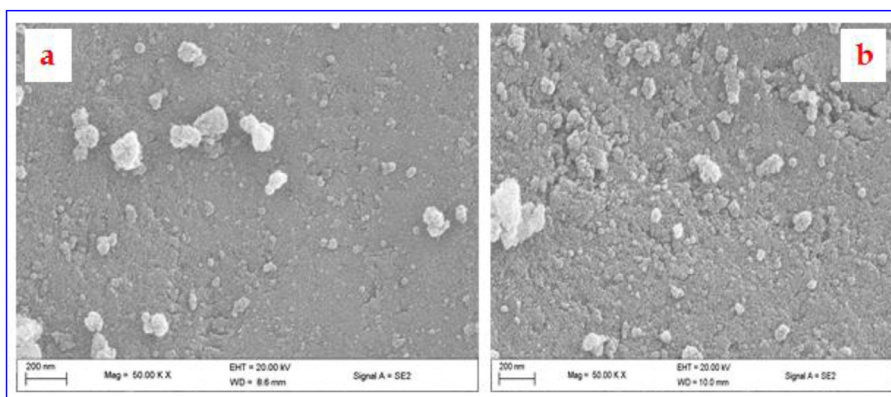


Fig. 2. Particulate depositions of SPM tobacco (a) and (b) ES1 from tobacco burning at an associated magnification of X50000 at 200 nm.

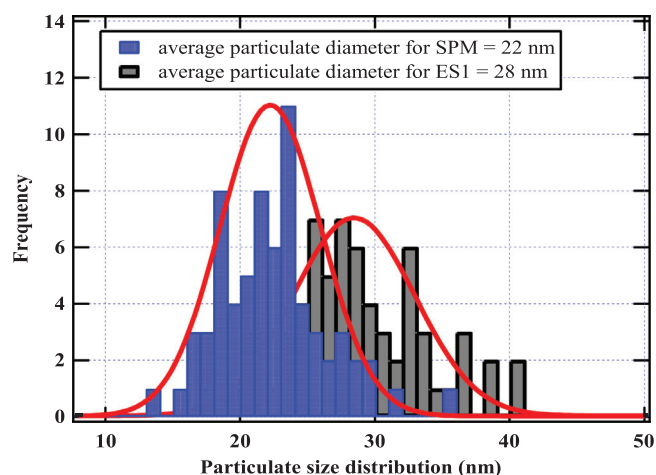


Fig. 3. Particulate distribution from tobacco burning of SPM cigarette (blue bars) and ES1 cigarette (black bars). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

a distribution curve of smoke size was then predicted using Igor ver. 5.0 graphing software for classification of cigarette smoke particulate emissions. A total of 80 spherical particulates were measured from each micrograph.

Results and discussion

The images presented in Figs. 2 were obtained at various scanning parameters; an extra high tension voltage (EHT) of 20.0 kV at various working distances (WD) as can be noted from the micrographs. The morphology of SPM tobacco (Fig. 2a) show particulate emissions of various sizes taken at an associated magnification of X50000. On the other hand, the morphology of ES1 cigarette show particulate emissions of various sizes (Fig. 2b).

The average size of particulate emissions from the thermal degradation of the two cigarettes was found to be ultrafine; 22 and 28 for SPM and ES1 tobacco cigarettes, respectively, (cf. Fig. 3) and can effectively be classified as ultrafine particulate matter. These particle sizes are filter independent because the smoke particulates did not pass through the cigarette filter and this affects the size of tobacco smoke particulates significantly. From a medical perspective, the ultrafine nature of tobacco smoke detected from this investigation is fatal if inhaled deeper into the lung surface. Because of the minute nature of these emissions, there is a possibility the particulates can penetrate into the blood stream and be carried along to the heart, thus initiating grave biological cell damage and heart diseases such as cardiac arrest, oxidative stress and cancer [31].

The ultrafine particulates emissions especially those from cigarette smoke are more harmful to human health than the large particulate emissions. The particulate matter having a diameter less than $2.5\mu\text{m}$ ($\text{PM}_{2.5}$), as is the case in this study are capable of by-passing the body's respiratory filters and penetrating deep into the lungs causing serious alveoli damage and irreversible injury to the lung macrophages [14]. Additionally, ultrafine particle emissions are associated with upper and lower respiratory damage as well as retardation of lung growth and rib cage malfunctions [32] thus, initiating malignant growth in the lungs and subsequently cancer of the lungs.

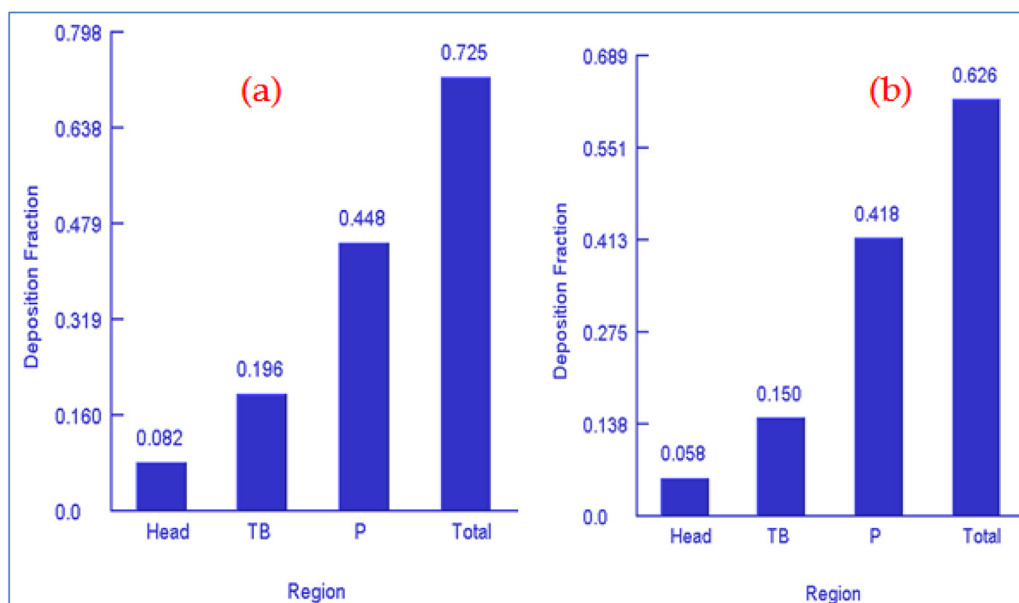


Fig. 4. Respiratory tract deposition fraction from the Multipath Particle Deposition (MPPD) model version 3.04, in the entire lung for particles with count median diameter (CMD) of (a) 22 nm and (b) 28 nm having a geometric standard deviation (GSD) of 1 nm.

Although exposure to $PM_{0.1}$ has been linked to reduced lung growth and lung function in children, the underlying biological mechanisms by which this health problem occurs is yet to be understood [33]. The two cigarettes (SPM and ES1) investigated in this work have a PM diameter of less than $0.1 \mu m$ (cf. Fig. 3). Therefore, inhaling the cigarette smoke from tobacco burning is detrimental to the biological health of cigarette smokers. Ultrafine particulates are also capable triggering the production of reactive oxygen species (ROS) which are well established precursors for severe oxidative stress within cells through the formation of oxidized cellular macromolecules, including lipids, proteins, and DNA [34].

Simulated breathing scenario and particulate deposition in the respiratory tissues

Dose estimation of particulate matter is a central component in quantitative risk assessment and the possible toxicological impact it inflicts on the organism [22]. In order to predict the deposition fractions of particulate matter in the lung and the lobar tissues, the Multipath Particle Deposition (MPPD) model version 3.04 was used to augment the experimental data reported in this work. An oronasal-mouth breather scenario was assumed in the MPPD model [23]. From the data presented in Figs 4 and 5, it is evident that the clearance of particulate matter from the respiratory tract is defined by the particle diameter referred to as the count median diameter (CMD). For instance, the pulmonary deposition fraction for particles with a CMD of 22 and 28 nm having a geometric standard deviation (GSD) of 1 nm, and a default standard density of $1 g/cm^3$, and an estimated concentration of $1 g/m^3$ was predicted to be 0.448 and 0.418. This implies retention in excess of 7% for geometric particles of 22 nm in diameter in comparison to geometric particles of 28 nm in diameter. In the entire lung system, an excess of about 13% for particulates of 22 nm geometric diameter were retained in the lung tissues.

The aim of this simulation is to predict the dosimetry of inhaled particles and establish regions in the respiratory tract where toxicity is likely to occur. It predicts details regarding the fractionation of particulates as they diffuse through the respiratory system. Tracheobronchial (TB) fractional deposition was estimated at 0.196 and 0.150 for ultrafine particulates according to Fig. 4(a) and (b), respectively.

Based on simulation of respirable particulates determined experimentally in this work, the five pulmonary lobes - left upper [LU], left lower [LL], right upper [RU], right middle [RM], and right lower [RL] showed estimated deposition fractions of 0.098, 0.194, 0.097, 0.049, and 0.0193, respectively for particles with a CMD of 22 nm (Cf. Fig. 5a). The corresponding deposition fraction for particulates of CMD 28 nm is shown in Fig. 5b. These findings suggest that the ultrafine particulates of 22 nm geometric diameter have a high retention in the respiratory system as compared to the fairly larger particulates of 28 nm CMD.

Tar and char yields from tobacco burning

Whereas the distribution of tobacco char with temperature has been reported in literature [35,36], very little information on the distribution of tobacco tar from mainstream cigarette smoke with temperature has been documented. Therefore this study is of significant interest. SPM tobacco cigarette yielded more tar (Fig. 6a) than ES1 tobacco cigarette (Fig. 6b). The tar

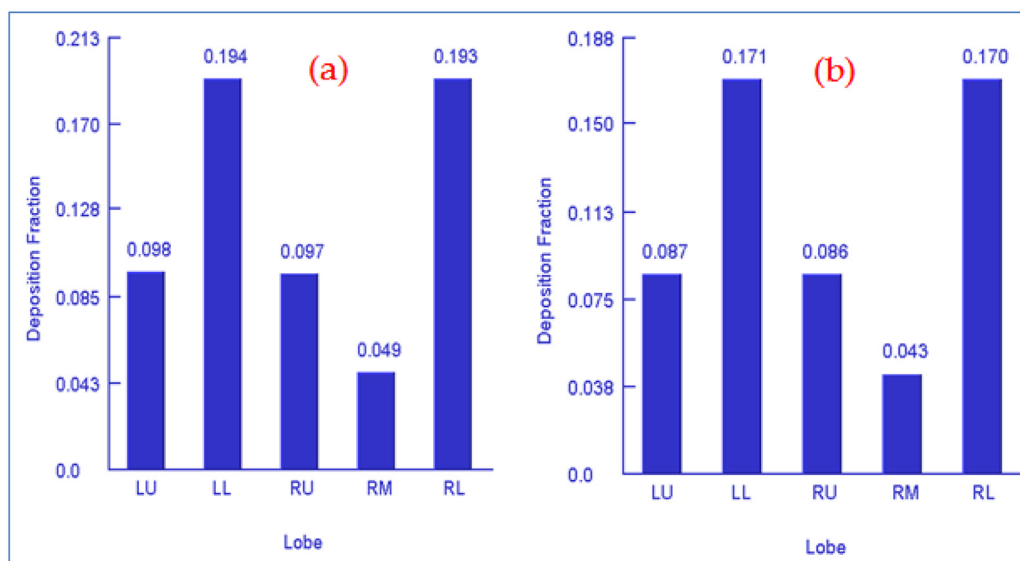


Fig. 5. Respiratory tract deposition fraction from the Multipath Particle Deposition (MPPD) model version 3.04, in lobar regions for particles with count medium diameter (CMD) of (a) 22 nm and (b) 28 nm having a geometric standard deviation (GSD) of 1 nm.

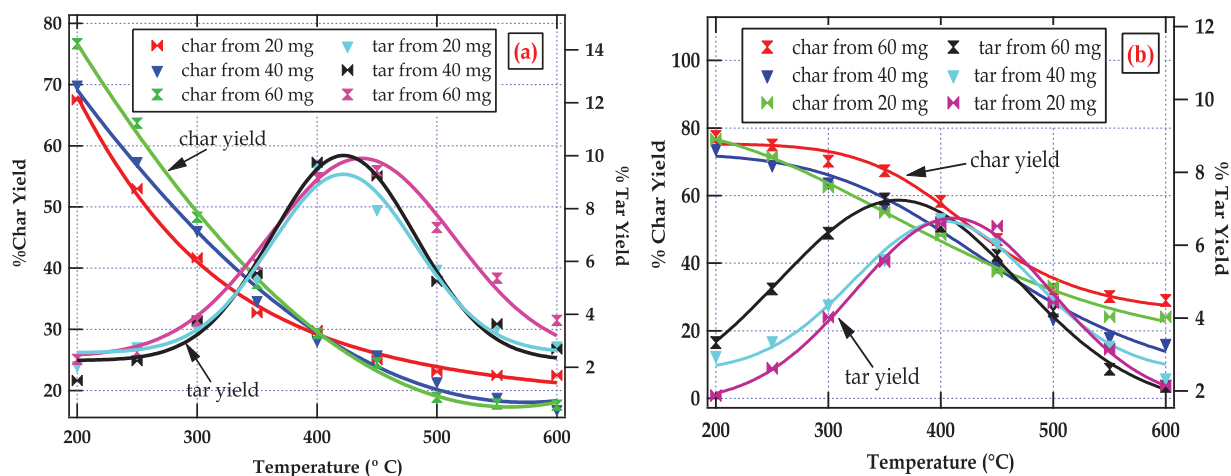


Fig. 6. The char and tar yield distribution at various pyrolysis temperatures for (a) SPM cigarette and (b) ES1 cigarette.

formation with temperatures for the two cigarettes are however slightly different. For SPM tobacco cigarette the maximum yields of tar were observed between 400 and 500 °C while for ES1 tobacco cigarette the maximum tar yields occurred between 300 and 450 °C (cf. Fig. 6). More recently, we investigated the char yields with temperature versus molecular products (components of tar) for various tobacco components and found to be consistent with the data reported in this study [35].

Evidently from Fig. 6, the mass loss due to the thermal degradation of SPM tobacco cigarette is significantly high between 200 and 400 °C (~42% on average). In the same temperature range (200 and 400 °C), the mass loss for ES1 tobacco cigarette was on average ~25%. Conversely, the tar yield within the same pyrolysis temperature range for the two tobacco cigarettes were similar ~25% on average. These results are remarkably interesting and imply one possibility – ES1 tobacco cigarette releases more gaseous by-products such as water vapor, methane, CO₂ etc. which are difficult to condense as tar. This observation may be a consequence of various additives introduced to tobacco during cigarette manufacture, tobacco growing conditions, and the nature of tobacco.

To obtain a better resolution of the inherent differences between the burning of the two tobaccos (SPM and ES1), the tar and char yields of the two cigarettes at 400 °C were selectively analyzed and presented in Table 1. It is interesting to note that the tar and char yields corresponding to every cigarette do not change significantly with the mass of tobacco used. This observation is consistent with literature data that char yield is independent of the mass of biomass material pyrolyzed [36]. Nonetheless, SPM cigarette generally gives low yields of char in the entire pyrolysis temperature range compared to

Table 1

Comparison of tar and char yields from the burning of SPM and ES1 tobacco at 400 °C.

Mass of tobacco used (± 2 mg)		60	40	20
SPM tobacco	% Tar yield	9.17 ± 1.2	9.75 ± 0.8	9.54 ± 0.6
	% Char yield	29.8 ± 2.2	28.5 ± 2.2	29.3 ± 2.2
ES1 tobacco	% Tar yield	6.53 ± 0.5	6.75 ± 0.4	6.6 ± 0.8
	% Char yield	48.3 ± 2.2	52.5 ± 3.4	53.3 ± 2.8

ES1 cigarette (cf. Fig. 6 and Table 1). On the other hand, the tar yield of SPM cigarette is slightly higher than the tar yields from ES1 cigarette. Based on this observation, it may be implied that ES1 cigarette has a higher concentration of molecular reaction products than SPM cigarette and probably more toxic. Therefore, unlike char, the amount of cigarette smoke released by burning a given cigarette is dependent on the nature of the cigarette brand [37].

As measured in our laboratory, the average mass of a commercial cigarette contains tobacco of about 650 ± 10 mg on average. Therefore, if approximately 25% of the tar component is generated in the entire smoking temperature range, then about 160 mg of tar is inhaled by the smoker in mainstream cigarette smoking. Nonetheless, in presence of a catalytic filter to trap some of the tobacco tar as reported in literature [3], the inhaled tar is estimated at 13% per cigarette and may suggest that approximately 85 mg of tar is puffed in by a cigarette smoker. A heavy smoker, for example, smoking estimated 20 cigarettes sticks per day may end up depositing ~ 1700 mg (1.7 g) of cigarette tar on his lung surface. These findings, while startling, may explain why most cigarette smokers are frail and have short-life spans because of the toxic tar component deposited in their body tissues. However, it is important to note that the experimental conditions applied in this study were an estimate of the actual human smoking conditions designed to assess how much tar may be deposited in the human lung. Remarkably, our results are approximately consistent with what has been reported elsewhere in literature [8].

Conclusion

This study has suggested that various masses of tobaccos from different cigarettes may yield different amounts of smoke condensate (tar) depending on the nature of tobacco, tobacco additives and tobacco growing conditions. Generally, high yields of tar were produced between 300 and 400 °C. This temperature region if avoided during cigarette manufacture can result to health benefits to the cigarette smoking community. Moreover, this work has established that tobacco smoke particulate emissions from tobacco burning are ultrafine implying that if inhaled, these particulates can enter into the blood circulation system to cause serious biological harm including cardio-pulmonary death, oxidative stress, nervous breakdown, chronic coughs, and cancers. Deposition fractions of particles for different body tissues of the human respiratory tract were calculated and found that the pulmonary retained the highest fraction of particulates. Furthermore, in the lobar regions; the LL and the RL had the highest deposition fraction which is consistent with various literature surveys on breathing and inspirational clearance of inhalants by the respiratory system.

Authors' contributions

TK designed the project. AJ prepared tobacco samples and conducted pyrolysis experiments under the direction of JK and TK, and wrote the first draft of the manuscript. VN conducted SEM analysis and made critical suggestions towards the improvement of the manuscript. JK conducted MPPD simulations, and critically reviewed the manuscript. All authors read and approved the final manuscript.

Conflict of interests

The authors declare they have no competing interests.

Funding

The authors are grateful to the [National Research Foundation](#) (South Africa) grant #103979 and Egerton University grant #EU/RE/DVC/072 for co-funding this research.

Acknowledgment

The authors are thankful to Caren Kurgat (Ms) for reviewing this article and making critical inputs that improved the scientific quality of the article. Applied Research Associates (ARA), Inc., Raleigh, NC, are greatly appreciated for providing us with MPPD software used in the respiratory simulation of deposition and clearance of particulates from tobacco burning.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:[10.1016/j.sciaf.2018.e00004](https://doi.org/10.1016/j.sciaf.2018.e00004).

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