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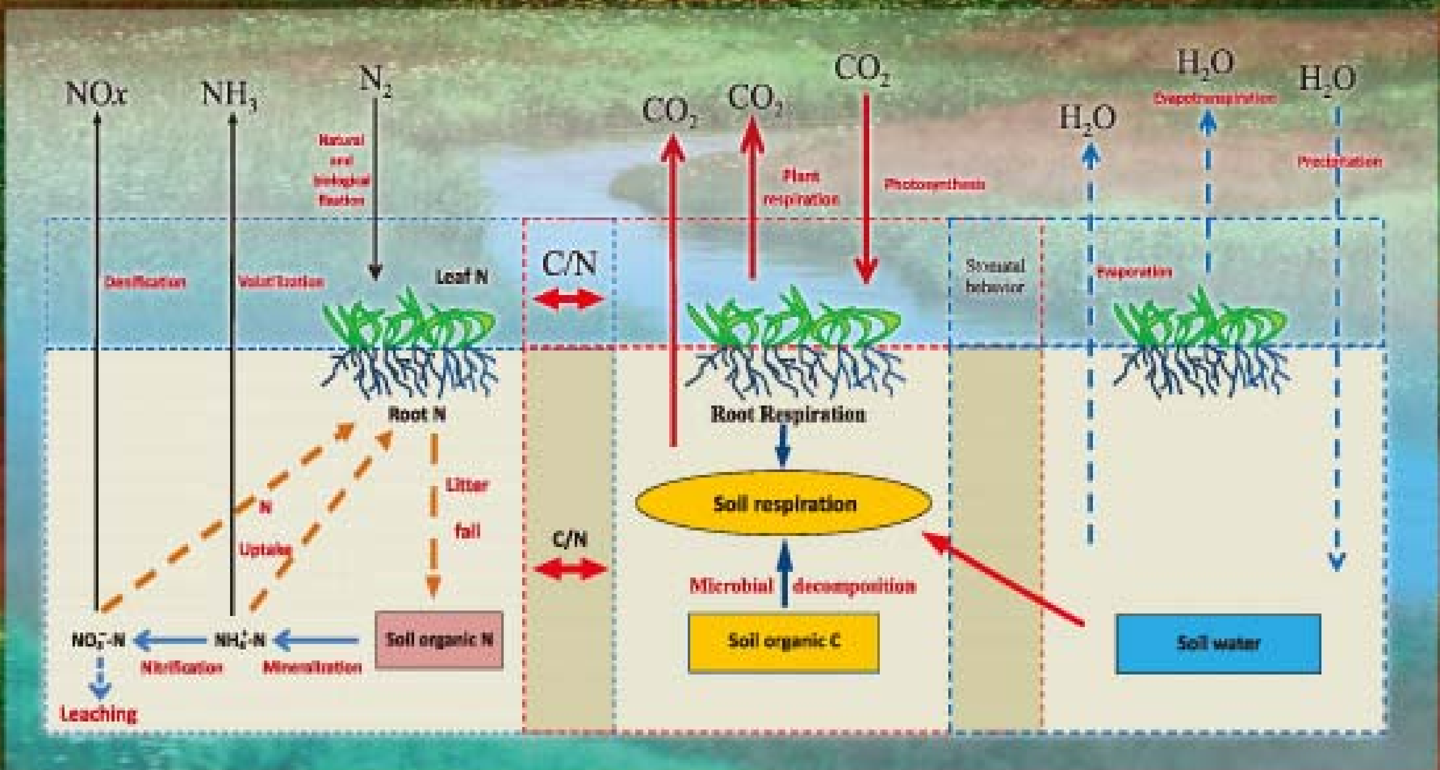

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Characterization of particle size distribution of mainstream cigarette smoke generated by smoking machine with an electrical low pressure impactor

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ABSTRACT

Cigarette smoking is a particle-related exposure. Studying the characteristics of the particle size distribution of cigarette smoke can aid in providing knowledge of smoke aerosol attributes. We used an electrical low pressure impactor (ELPI) to measure the particle size distribution of mainstream cigarette smoke generated by a smoking machine and provided a continuum of particle sizes of cigarette smoke from a whole cigarette. The results showed that the aerodynamic diameters (D, geometric mean of a channel) of particles ranged from 0.021 to 1.956 μm, and the number concentrations were on the order of 10^5–10^9 cm^{-3} for different sizes of particles. The particle number of the size category below 0.1 μm approximated that of the category 0.1–2.0 μm, and the particles in the size category of 0.1–2.0 μm contributed extremely heavily to total particulate mass. In addition, the results with small samples indicated that the tar yields normalized per milligram of nicotine showed an approximately linear increase with increasing concentration of total particles.

Introduction

Cigarettes are the most-used combusted tobacco products by far, and the burning of tobacco during smoking causes smokers to be exposed to thousands of toxicants. The health effects of cigarette smoking have been of more and more concern to the public and the government (FSPTCA, 2011; IOM, 2001). Mainstream cigarette smoke, which emerges from the butt end of a puffed cigarette, is a dynamic and complex aerosol composed of more than 5000 chemical constituents (Rodgman and Perfetti, 2008). The majority of constituents exist in the particulate phase and the minority is found in the gas vapor phase, while some constituents dynamically distribute in both phases. The chemical compounds, formed by pyrolysis and distillation during tobacco combustion, coagulate and form various sizes of particles which are small enough to be inhaled and deposit in the respiratory tract and the lung of smokers. The particles in smoke aerosol have been reported to play important roles in smoking-related diseases, such as lung cancer, chronic obstructive pulmonary disease (COPD) and cardiovascular disease (US DHHS, 1989; IARC, 2004; Sherman, 1991; Yoshida and Tuder, 2007).

Cigarette smoking is a particle-related exposure (Sangani and Ghio, 2011). The health consequences of smoking are mainly attributable to inhalation of particles. The particle size distribution of cigarette smoke aerosol is an important parameter in predicting the deposition of inhaled particles in different regions of the smoker's airways. Studying the characteristics of the particle size distribution of cigarette smoke can aid in providing knowledge about smoke aerosol (Aldermana and Ingebrethsen, 2011; Anderson et al., 1989; Hinds, 1978; McCusker et al., 1983), understanding the retention and deposition of cigarette smoke particles in the respiratory tract (Bernstein, 2004;

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Kane et al., 2010), and performing modification of product design (Wayne et al., 2008). In addition, the measurements of particle size distribution associated with the data from smoke chemical analysis and biological assays can provide a comprehensive insight into the adverse effects of cigarette smoke and the health risk following smoking. Over the past decades, the tobacco industry has long been involved in harm reduction for tobacco products. Smoke aerosol particle size manipulation has been evaluated both as a means of controlling physical and sensory product attributes and as a possible approach to reducing health risk related to exposure (Wayne et al., 2008).

The differences in the measurements of particle size distribution of cigarette smoke, which have been reported previously, are attributed to differences in measurement methods. Hinds (1978) measured the aerodynamic size distribution of cigarette smoke by using an aerosol centrifuge and a cascade impactor, and observed that mass median diameter decreased from 0.52 to 0.38 μm with increasing dilution. Anderson et al. (1989) reported that mass median diameter measured by the electrical aerosol analyzer was smaller than in previous data. Recently, with the development of advanced aerosol analysis technology, some of the instruments used in the fields of aerosol measurement and sampling have been applied in combustion research, including cigarette smoke (Adam et al., 2009; Aldermana and Ingebrethsen, 2011; Becquemin et al., 2009; Kane et al., 2010). Kane et al. (2010) used an electrical low pressure impactor (ELPI) to investigate effects of smoking parameters on the particle size distribution and predicted airway deposition of mainstream cigarette smoke, and they determined that higher puff flow rates and reduced filter ventilation decreased the count median diameter of cigarette smoke. Aldermana and Ingebrethsen (2011) used a DMS500 fast particulate spectrometer to characterize the particle size distribution of mainstream cigarette smoke.

In this study, an ELPI was employed to measure the particle size distribution of mainstream cigarette smoke generated by a smoking machine. The purpose of this study was: (1) to provide a continuum of particle sizes of cigarette smoke from a whole-cigarette measurement, and (2) to compare the particle distribution between the size category below 0.1 μm and the category of 0.1–2.0 μm. In addition, the correlation between the concentration of particles and tar yields or nicotine yields was studied preliminarily.

1 Materials and methods

1.1 Cigarettes

Kentucky reference cigarettes 3R4F from the University of Kentucky (Lexington, Kentucky, USA) and seven brands of commercial cigarettes purchased from the Chinese market were used in this study. The cigarettes were conditioned at 22 ± 1°C and 60% ± 3% relative humidity for at least 48 hr before being smoked.

1.2 Mainstream cigarette smoke particle size distribution measurement

The particle size distribution of fresh smoke aerosol was measured with an ELPI (Dekati, Tampere, Finland). The ELPI is a real-time particle size spectrometer for real-time monitoring of aerosol particle size distribution (Keskinen et al., 1992). The main components of this instrument are a corona charger, 13 stage cascade low-pressure impactor and multichannel electrometer. The ELPI operates at 10 L/min air flow, and measures particle size ranging from 7 nm with filter stages to 10 μm.

Cigarettes were smoked by a Borgwaldt LM1 smoking machine (Borgwaldt KC, Hamburg, Germany) under the International Organization for Standardization (ISO) smoking regimen (35/60/2 without blocking of filter ventilation) (ISO, 2000). When a puff was generated, a sample of smoke passed through the axial diluter (Dekati) by means of a vacuum pump, and then the diluted smoke passed through the second diluter (Dekati), in which it was mixed with clean air. After being twice diluted, a sample of diluted smoke was introduced into the inlet of the ELPI for particle size distribution measurement (**Fig. 1**). Total dilution ratios between 600 and 1000 were achieved for whole system.

The charged particles in smoke aerosol collected on the 13 collection plates of the impactor according to their aerodynamic diameter were recorded on 12 electrometer channels. The measurement data were processed by ELPI XLS4.05 software. The particle size measured by impact was equivalent to the aerodynamic diameter of a particle. In this study, the aerodynamic diameter was addressed using D (geometric mean of a channel). The particle size range and D corresponding to each impactor stage are shown in **Table 1**.

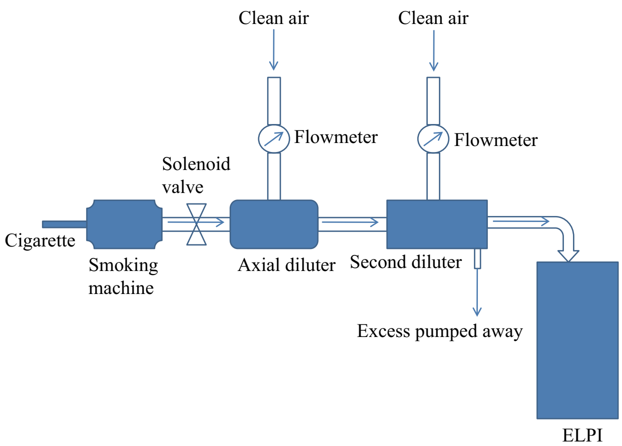

Fig. 1 Schematic diagram of the smoking machine and dilution system for the electrical low pressure impactor (ELPI) measurements.

Table 1 Particle size range and D corresponding to each impactor stage

Impactorst age	Particle size range (µm)	D (µm)
1	0.007–0.029	0.021
2	0.029–0.057	0.039
3	0.057–0.101	0.070
4	0.101–0.165	0.119
5	0.165–0.255	0.201
6	0.255–0.393	0.315
7	0.393–0.637	0.483
8	0.637–0.990	0.761
9	0.990–1.610	1.231
10	1.610–2.460	1.956
11	2.460–3.970	3.088
12	3.970–10.150	6.285

1.3 Chemical analysis

The yields of tar and nicotine were determined according to ISO 4387 (ISO, 2000).

1.4 Statistical analysis

The data were expressed as mean ± SD. One-way ANOVA analysis was applied to compare the results from experiments. The value of $P < 0.05$ was considered as statistically significant.

2 Results

2.1 Particle size distribution and concentration of mainstream cigarette smoke from a whole cigarette

Figure 2 shows the particle size distribution and concentration of mainstream cigarette smoke from 3R4F reference cigarettes generated by the smoking machine. The aerodynamic diameters (D) of particles ranged from 0.021 to 1.956 µm. For different sizes of particles, the number concentrations were on the order of 10^5–10^9 cm^{-3}. **Figure 3a** shows the particulate mass concentration of smoke aerosol, and there were large differences in the particulate mass concentrations among different size particles. The mass of particles of 0.021, 0.039, 0.070 or 0.119 µm was less than 1% of total particulate mass, respectively. The respective contribution to total particulate mass for particles of 0.315, 0.483 or 0.761 µm was all more than 10%. And the mass of particles of 0.201, 1.231 or 1.956 µm was less than 10% but more than 1% of total particulate mass, respectively (**Fig. 3b**).

Table 2 shows the comparison of the particle distributions between the size category below 0.1 µm and the category of 0.1–2.0 µm. Seven brands of commercial cigarettes and 3R4F reference cigarettes were used for analysis. The “Tar” yields of these cigarettes as labeled on the packages ranged from 5 to 12 mg/cigarette. The total particulate mass concentrations of mainstream cigarette smoke from these cigarettes were between 27.88 and 48.38 µg/cm^3, and the total particle number concentrations of smoke aerosol were between 1.34×10^9 and 2.73×10^9 cm^{-3}. There was no significant difference in particle number between the size category below 0.1 µm and the category of 0.1–2.0 µm. However, there was significant difference in particulate mass between the two categories of particles ($P < 0.01$), and the particles in the size category of 0.1–2.0 µm contributed extremely heavily to total particulate mass.

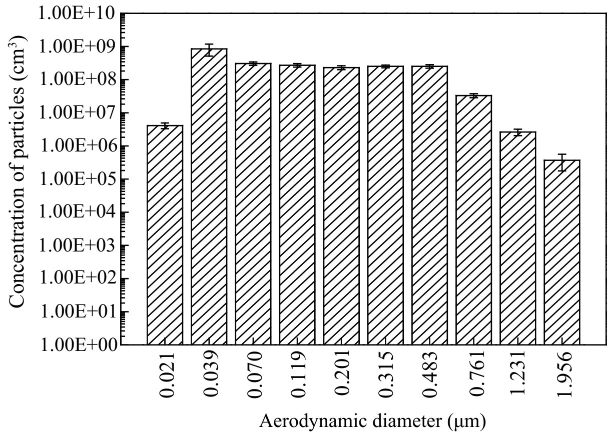

Fig. 2 Particle size distribution and concentration of mainstream cigarette smoke from 3R4F reference cigarettes. Mainstream cigarette smoke from 3R4F reference cigarettes generated by smoking machine was measured with the ELPI. The measurements were carried out in six independent experiments, and the data are expressed as mean ± SD.

2.2 Particle size distribution and concentration of puff-by-puff mainstream cigarette smoke

Figure 4 shows the particle size distribution and concentration of puff-by-puff mainstream cigarette smoke from 3R4F reference cigarettes generated by the smoking machine. The 3R4F reference cigarette was smoked seven puffs under the ISO smoking regimen. For every puff of smoke, the aerodynamic diameters of particles ranged from 0.021–1.956 µm. The particle size distribution and concentration of puff-by-puff smoke were similar to those of smoke aerosol generated from a whole cigarette. There was no difference in the characteristics of particle size distribution among puffs. The particulate mass concentration of every puff of smoke was similar to that of the whole-cigarette smoke aerosol, as was the contribution of different size particles to the total particulate mass for every puff (**Fig. 5**).

2.3 Correlation between the concentration of particles and tar yields or nicotine yields

Seven brands of commercial cigarettes and 3R4F reference cigarettes were used for analysis. There was no linear

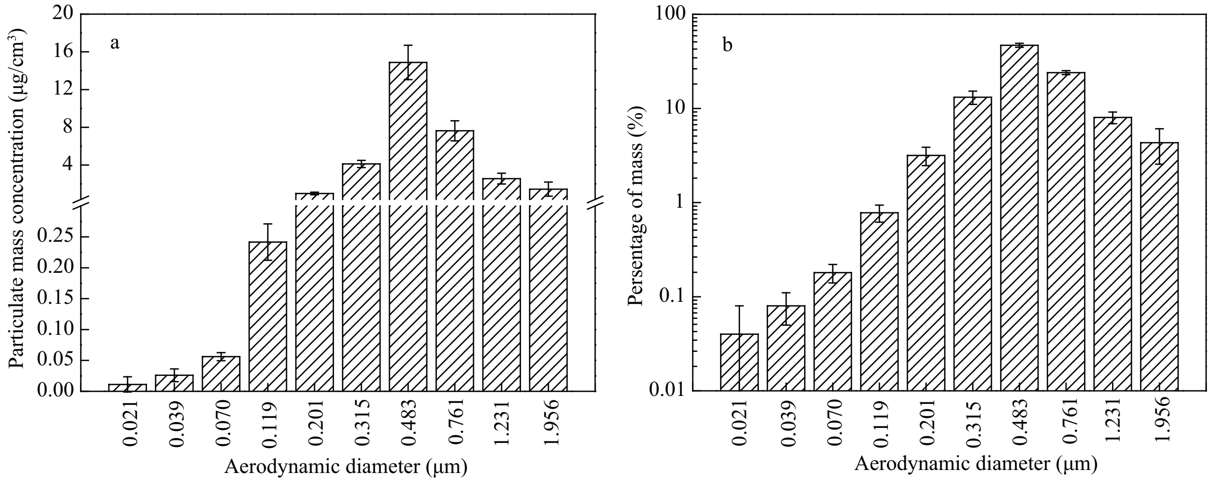

Fig. 3 Particulate mass concentration of smoke aerosol (a) and the contribution of different size particles to total particulate mass (b). Mainstream cigarette smoke from 3R4F reference cigarettes generated by smoking machine was measured with the ELPI. The measurements were carried out in six independent experiments, and the data are expressed as mean ± SD.

Table 2 Comparison of the particle distribution between the size category below 0.1 μm and the category of 0.1–2.0 μm

Cigarette ID	"Tar"# (mg/cigarette)	Replicates (*n*)	Particulate mass concentration (μg/cm³)	Concentration of particle (× 10^9 cm^{-3})	Percentage of number (%)		Percentage of mass (%)	
					< 0.1 μm	0.1–2.0 μm[a]	< 0.1 μm	0.1–2.0 μm[b]
3R4F	9.5	6	37.71 ± 3.73	2.19 ± 0.28	51.8 ± 7.8	48.2 ± 7.8	0.3 ± 0.1	99.7 ± 0.1
DB (blended type)	10	5	37.77 ± 1.74	2.32 ± 0.22	47.1 ± 7.2	52.9 ± 7.2	0.3 ± 0.0	99.7 ± 0.0
ZHNH (blended type)	10	5	33.03 ± 2.59	1.87 ± 0.21	45.8 ± 11.8	54.2 ± 11.8	0.3 ± 0.0	99.7 ± 0.0
TF (Virginia type)	12	6	40.36 ± 4.55	1.83 ± 0.40	43.5 ± 10.7	56.5 ± 10.7	0.1 ± 0.0	99.9 ± 0.0
WBL (blended type)	12	6	37.82 ± 3.86	2.48 ± 0.71	52.1 ± 13.3	47.9 ± 13.3	0.2 ± 0.1	99.8 ± 0.1
ZHH (Virginia type)	12	3	48.38 ± 4.86	1.87 ± 0.33	34.4 ± 0.8	65.6 ± 0.8	0.1 ± 0.0	99.9 ± 0.0
CHBS (Virginia type)	5	3	31.21 ± 5.83	1.34 ± 0.25	35.0 ± 0.9	65.0 ± 0.9	0.1 ± 0.0	99.9 ± 0.0
ZHNH (blended type)	5	3	27.88 ± 7.55	2.73 ± 0.37	67.2 ± 2.4	32.8 ± 2.4	0.4 ± 0.0	99.6 ± 0.0

The data were expressed as mean ± SD.

The "Tar" values are as labeled on packages of cigarettes.

[a] No significant difference between particle number of size category < 0.1 μm and 0.1–2.0 μm (P = 0.29), [b] significant difference between particulate mass of size category < 0.1 μm and 0.1–2.0 μm (P < 0.01).

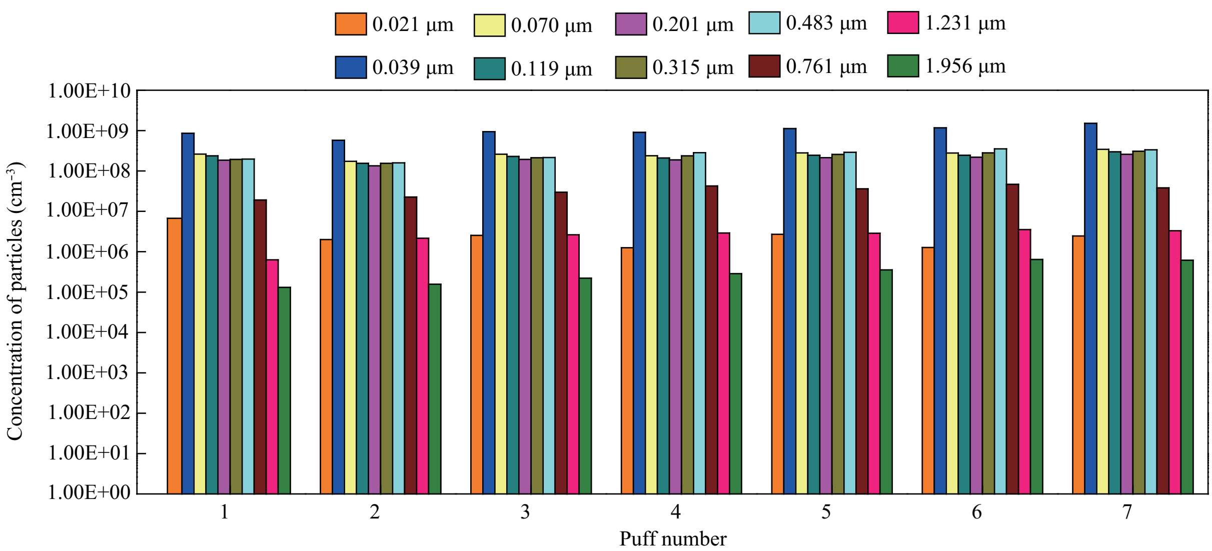

Fig. 4 Particle size distribution and concentration of puff-by-puff mainstream cigarette smoke.

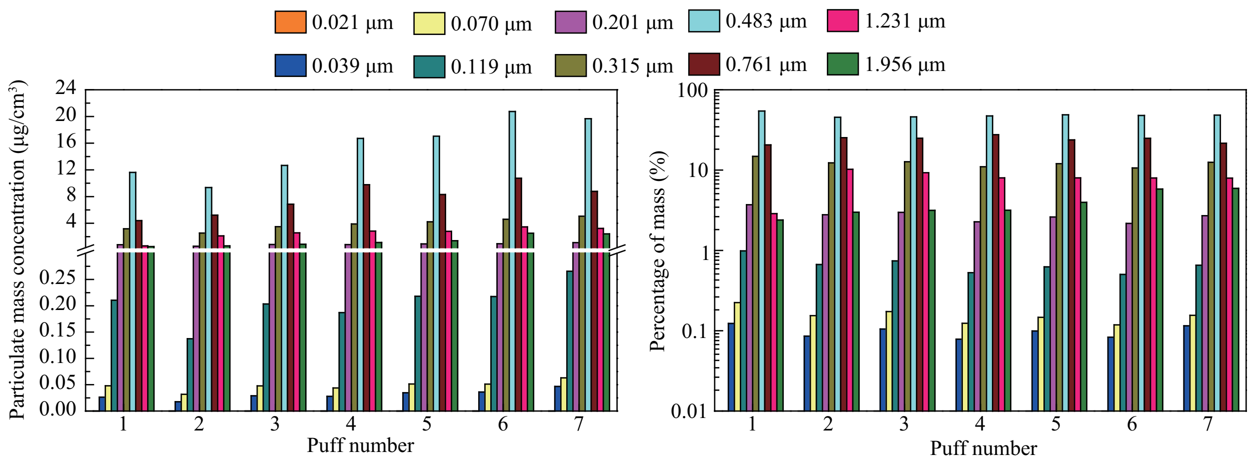

Fig. 5 Particulate mass concentration of puff-by-puff mainstream cigarette smoke (a) and the contribution of different size particles to total particulate mass for every puff (b).

correlation between the concentration of total particles and tar yields per cigarette (**Fig. 6a**), nor between the concentration of total particles and nicotine yields per cigarette (**Fig. 6b**). When tar yields were expressed on a per-mg of nicotine basis, the tar yields showed an approximately linear increase with increasing concentration of total particles (R^2 = 0.684) (**Fig. 6c**).

3 Discussion

Cigarette smoke is a complex aerosol comprised of a particulate phase and gas vapor phase. Particles in smoke, carrying toxicants, are inhaled into the respiratory tract and cause smoking-related diseases. The particle size distribution of smoke aerosol is related closely to the chemical composition and biological effects of particulate matter. The ELPI is a real-time particle size spectrometer designed at the Tampere University of Technology for measurement of aerosol particle size distribution (Keskinen et al., 1992). This instrument can monitor airborne particle size distributions in the range of 7 nm with filter stages to 10 μm, and is qualified for different applications, such as automotive exhaust emissions (Liu et al., 2011), pharmaceutical studies (Ali, 2010) and combustion research (Nussbaum et al., 2009). In this study, the particle size distribution of mainstream cigarette smoke generated by a smoking machine was measured by an ELPI. The results showed that the aerodynamic diameters (D) of particles ranged from 0.021 to 1.956 μm, and the number concentrations were on the order of 10^5–10^9 cm^{-3} for different sizes of particles. The total particle number concentration was on the order of 10^9 cm^{-3}, which is consistent with previous reports (Bernstein, 2004; Kane et al., 2010). Large differences in the particulate mass concentration were observed among different size particles.

An interesting result was found when all particles in smoke were classified into two categories, the size category below 0.1 μm and the category of 0.1–2.0 μm. There was no significant difference in particle number between the two categories, while the particulate mass of particles in the category of 0.1–2.0 μm was more than 99% of total particulate mass. These results were consistent in all

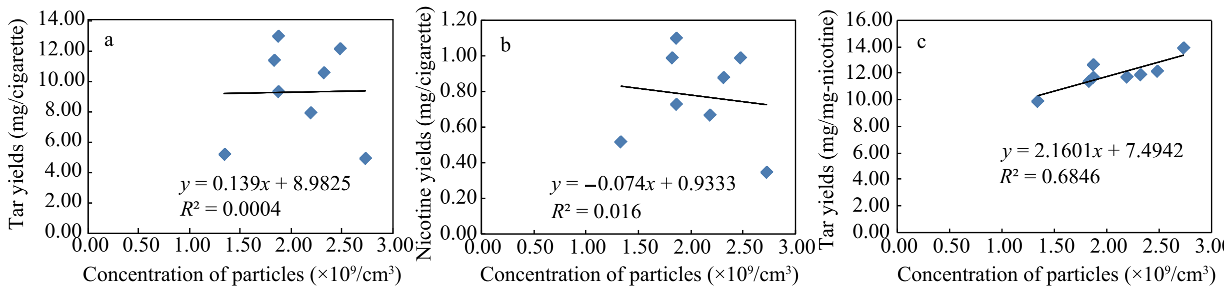

Fig. 6 Correlation between the concentration of particles and tar yields or nicotine yields. (a) correlation between the concentration of total particles and tar yields per cigarette; (b) correlation between the concentration of total particles and nicotine yields per cigarette; (c) correlation between the concentration of total particles and tar yield on a per-mg of nicotine basis. The concentration of particles as well as the values of tar or nicotine yields is the mean value for each brand of cigarettes. The correlation analysis was conducted using Microsoft Office Excel 2007.

test cigarettes. Particles smaller than 0.1 μm are labeled ultrafine particles, and particles between 0.1 and 2.5 μm are termed fine particles (Wichmann et al., 2000). Wichmann et al. (2000) concluded that both fine particles (represented by particle mass) and ultrafine particles (represented by particle number) showed independent effects on mortality at ambient concentrations. Epidemiological and toxicological studies show that inhalation of fine and ultrafine particles may cause adverse health effects, which might be related to factors including mass, surface area and number concentration of particles (Brouwer et al., 2004). The data obtained from this study showed that the smoke aerosol was composed of fine and ultrafine particles, and the particulate mass was dominated by fine particles and the contribution to particle number was similar for both fine particles and ultrafine particles.

The particulate matter in cigarette smoke carries a variety of chemical constituents, which form the tar, as well as nicotine and water. The study of tar has been generally focused on its toxicological effects, and nicotine is the addictive component of cigarette smoke. In the present study, we carried out a preliminary study on the correlation between the concentration of particles and tar yields or nicotine yields. There was no linear correlation between the concentration of total particles and tar yields per cigarette or with nicotine yields per cigarette. McCusker et al. (1983) previously reported that the number concentration of smoke particles from some “low” tar cigarettes was found to be similar to that of some “medium” tar cigarettes. The World Health Organization Study Group on Tobacco Product Regulation (TobReg) recommends that toxicant yields be normalized per milligram of nicotine for the regulation of tobacco products. The purpose of this normalization is to shift the interpretation of measurements away from the quantity of smoke generated per cigarette and the misleading use of the machine-measured yields as data of smoker exposure and risk (TobReg, 2008). Reporting results on a per nicotine basis represents theoretically a calculation to correct for a smoker’s nicotine intake based on the assumption that consumers smoke cigarettes to titrate their blood nicotine levels (Johnson et al., 2009). Hence, we normalized tar yields on a per-mg of nicotine basis, and found that the shifted tar yields showed an approximately linear increase with increasing concentration of total particles. The results suggest that decreasing the concentration of total particles in cigarette smoke might reduce the tar yields on a per-mg of nicotine basis. Certainly, the small number of test samples is a limitation for this result, so there needs to be further work with a large number of brands of cigarettes to confirm the above results in the future. In addition, we recommend further analysis of the chemical characteristics of smoke particulate matter so that the physicochemical properties of smoke aerosol can be explained explicitly.

4 Conclusions

The number concentrations for different size of particles in cigarette smoke generated by a smoking machine are on the order of 10^5–10^9 cm^{-3}, and there are large differences in the particulate mass concentration among different size particles. The particle number of the size category below 0.1 μm approximates that of the category of 0.1–2.0 μm. However, the particles in the size category of 0.1–2.0 μm contribute extremely heavily to total particulate mass. In addition, this study with small sample size indicates that the tar yields normalized on a per-mg of nicotine basis show an approximately linear increase with increasing concentration of total particles. In the present study, all particle size measurements were performed under the ISO machine-smoking regimen. It is well known that no machine-smoking regimen can represent all human smoking behaviors. However, machine-smoking testing is useful to characterize cigarette emissions for product design and regulatory purposes. Therefore, the data obtained from this work could provide a referenced insight into the physical properties of fresh cigarette smoke aerosol. The further study of chemical characteristics and biological responses of particles of different sizes associated with the physical properties of cigarette smoke will provide information for a comprehensive understanding of the mechanisms of smoking-related diseases.

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