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MATHEMATICAL MODELING OF CIGARETTE SMOKE PARTICLES DYNAMICS IN HIGH AIR CHANGE RATE CHAMBERS LIKE CARS

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Abstract. Dynamics of cigarette smoke particles in high air change rate chambers like cars is investigated and the RSP concentration during and after smoking the cigarette is predicted in this study. This model is based on the mass balance equation. The generation and removal rate of RSP for various particle sizes are specified from the cigarette emission profile, ventilation rate, filtration efficiency, coagulation of particles and deposition rate on surfaces. The cigarette emission profile serves as the main source term in the mass balance equation. Ventilation rate and filtration of the cabin are considered as sinks for emitted particles due to the cigarette smoking in cars. These effects together with the mechanism of deposition on the interior surfaces are the main sinks for cigarette smoke particles in high surface-to-volume ratio chambers like cars. The coagulation is taken into account as a sink for smaller particles and as a source for larger particles. The conducted experiments on the PM_{2.5} concentration level after smoking the cigarettes in cars are summarized and the smoking scenarios of the reported experiments are simulated in this study. In most of experimental studies in cars, the total PM_{2.5} concentration is measured and reported, thus, the predicted concentration of each size section is integrated over the all range of particle sizes up to 2.5 μm to obtain a value comparable with the total PM_{2.5} concentrations in the literature. Good agreement between the results of the present model and the experimental data is reached. The results of the current model are also compared with the presented mathematical model in the literature on the RSP concentration in cars (e.g. USEPA Indoor Air Quality Model). The similar trend of RSP concentration are observed between the results of the different models and the minor quantitative discrepancies are most likely due to different cigarette particle emission rate being incorporated in the various models.



1 INTRODUCTION

Environmental tobacco smoke (ETS) emission is a major source of Respirable Suspended Particles (RSP) in enclosed areas. ETS consists of a mixture of large number of various substances which are distributed as particles, vapors, and gasses in the environment as the results of burning of tobacco products [1]. RSP are those particles which can penetrate deep and deposit on the lungs of humans and their aerodynamic diameters are usually less than $2.5\mu\text{m}$ ($\text{PM}_{2.5}$). Until now, at least 4700 components of ETS have been recognized, and the number of unknown constituents has been estimated to be as high as 100,000 [2]

Many studies in the past decades have measured and modeled the pollutants of cigarettes in different locations, including public, residential, and commercial places. Public lounges, restaurants, taverns, bars, hospitals, and offices are among the most common locations in past studies [3-7].

In cars, because of the limited space in which the cigarette smokes disperses, the concentration of cigarette pollutants is expected to be higher than in other places. An experimental investigation that was recently reported by the Ontario Tobacco Research Unit [8], showed that smoking just a single cigarette in a car can lead to high level of tobacco smoke concentration that exceeds by several times the amount of tobacco smoke concentration found in the smokiest bars and restaurants. High RSP concentration after smoking a cigarette in cars is the main threat on health of nonsmokers especially children.

The most of the models presented for predicting the pollutant concentration in enclosed areas are based on the mass balance equation. Nazaroff and Case [8] simulates the evolution of particle size distributions and predicts the concentration of different particle sizes as a function of time. In this aerosol dynamics model (MIAQ4), many factors like direct ETS particle emissions, inter-zonal mixing, ventilation, filtration, coagulation, and deposition onto indoor surfaces, have been accounted for each size section of the particles. Ott et al. [9] also developed the sequential cigarette exposure model (SCEM) for calculating the pollutant concentration time series in well-mixed environments.

In this study, the behavior of cigarette smoke particles was simulated in chambers with high air change rate like cars. The concentration of the respirable particles was predicted for different car status, windows status and ventilation modes as a function of time. The range of the particle diameter in cigarette emission profile was divided into 8 size sections and the concentration was calculated for each size section, separately.

2 EXPERIMENTAL STUDIES IN CARS

By increasing the prevalence of cigarette smoking in cars, assessing the threats of this habit on the health of nonsmokers persuade researchers to measure the pollutant concentration in cars after smoking cigarettes. Table 1 summarizes all the conducted experiments on the $\text{PM}_{2.5}$ concentration level after smoking cigarettes in cars from 4 papers. Different ventilation scenarios and windows positions of cars lead to different quantity of RSP concentration. These varieties can be caused by different procedure of each experiment and is dependent to other factors like

measuring the sidestream or sidestream plus mainstream, time between the puffs and the type of the cigarette.

Table 1: Reported experimental data in the literature on the PM_{2.5} particle concentration in cars after smoking the cigarettes.

Car (Volume)	Source	Conditions	ACH (h ⁻¹)	SS or SS+MS	t _{cig} (min)	t _{sm} (min)	Max PM _{2.5} (µg/m ³)	Mean PM _{2.5} (µg/m ³)
1985 Mazda 626 4-door Sedan (3.7 m ³)	Ott et al (1992)	20 mph, windows closed, AC set on recycling mode	7.27	SS+MS	8 ^a	72	2723	841
1991 Honda Civic sedan (2.2 m ³), 1986 Toyota Tercel (2.6 m ³), 2005 Honda Civic sedan (2.3 m ³)	Rees and Connolly (2005)	30-40 mph, four half (25 cm) open windows, AC off	-	SS+MS	5	15	51	27.8
		30-40 mph, one 5 cm open window near smoker, AC off	-	SS+MS	5	15	272	140
2005 Toyota Corolla compact (2.7 m ³)	Ott et al (2008)	20 mph, windows closed, fan off, AC off	1.9	SS	10 ^b	46	3035	1163
		parked, one open passenger's window, AC off	19.2	SS+MS	5.5	38.7	705	82.4
		20 mph, windows closed, AC max	3	SS+MS	5	27.2	3184	113
2005 Ford Taurus mid-size (2.4 m ³)	Ott et al (2008)	20 mph, one 3" open passenger's window, AC off	20.9	SS+MS	5.5	12.5	683	119
		20 mph, one fully open passenger's window, AC off	78.6	SS+MS	5	10.8	371	96.6
		20 mph, windows closed, AC regular	32.1	SS+MS	5.5	15	2389	529
		60 mph, windows closed, AC regular	38.6	SS+MS	5.5	14.5	1394	465
		60 mph, one 3" open passenger's window, AC off	56.4	SS+MS	5	9	608	119
		59 mph, windows closed, AC max	5.1	SS+MS	5.5	43	3808	658
		62 mph, windows closed, vent off, with recirculation	6	SS+MS	-	25.7	3212	1150
1999 Jeep Grand Cherokee Limited (3.0 m ³)	Ott et al (2008)	62 mph, windows closed, AC on, with recirculation	8.2	SS+MS	-	31	2828	1060
		62 mph, windows closed, AC on, no recirculation	28.4	SS+MS	-	14.5	1138	420
		60 mph, windows closed, AC on, no recirculation	23.4	SS+MS	-	25.7	1051	203.6
		60 mph, four 2" open windows, vent off, with recirculation	7.7	SS+MS	-	37.3	3104	627.6
18 different four-door cars (2.4-2.9 m ³ , avg. size = 2.6 m ³)	Sendzik et al. (2009)	parked, windows closed, AC off	-	SS+MS	7.5	25	5543	2920
		31 mph, windows closed, AC off	-	SS+MS	7.9	25	2530	1200
		31 mph, four open windows, AC off	-	SS+MS	5.9	25	54.3	24.2
		31 mph, one half (18 cm) open passenger's window, AC off	-	SS+MS	6.6	25	165	74.3
		31 mph, closed windows, AC on, fan set on medium speed	-	SS+MS	7.3	25	950	357

^a 3 cigarettes were smoked and the cigarette duration were 7 min, 12 s, 9 min and 8 min, 9 s, respectively.

^b cigarette is just smoldering in the car.

3 MATHEMATICAL SIMULATION

The most of the presented models in the literature for predicting the RSP concentration in enclosed areas are based on the mass balance equation and well-mixed assumption [8-9]. Assuming the uniform dispersing of the pollutant from a point source in large places like rooms and apartments can lead to considerable errors from the measurements until several minutes after the smoking finished. The basis of this modeling is given by a first-order differential equation as follows.

$$\frac{dC_i}{dt} = \dot{C}_{Generated,i} - \dot{C}_{Removed,i} \quad (1)$$

The generation and removing rate for each size section ($i=1-n$) is specified from cigarette emission profile, ventilation rate, filtration efficiency, coagulation of particles and deposition rate on surfaces.

3.1 Cigarette Emission Profile

The generation rate of RSP particles during the smoking is specified by cigarette emission profile for sidestream smoke or sidestream plus mainstream smoke. The ETS particle mass emissions size distribution for a burning cigarette has been reported by various investigators. The total mass emission rate of RSP particles of cigarette smoke was also reported by many researchers in the literature. Nazaroff and Klepeis [10] summarized the available conducted experiments on the total mass emission rate in ETS sources (cigar and cigarette), reported in 18 papers. Each study use specific method and setting for measuring the total mass emission rate of RSP particles. In the Fig. 1 and Fig. 2, the percentage of mass fraction for each size section in different reported experiments on sidestream and sidestream plus mainstream smoke are compared to each other. The percentage of each size section in two reported experiments on sidestream smoke profile are near to each other but for sidestream plus mainstream the experimental data show more variety on portion of each size section.

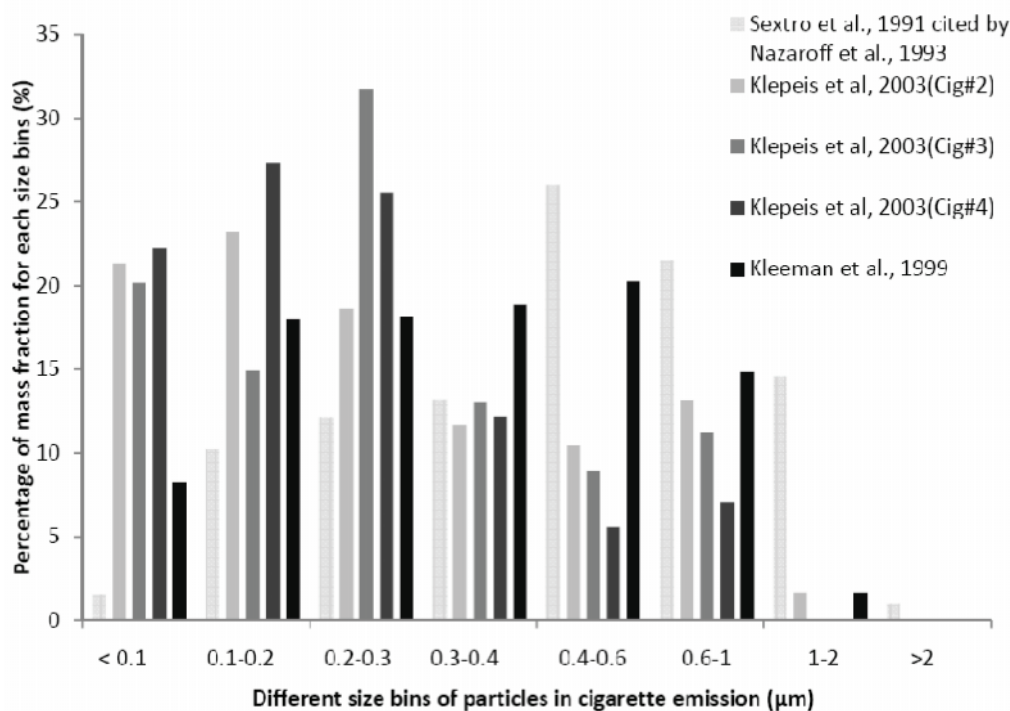


Fig. 1: Percentage of mass fraction for 8 size sections in different reported emission profile for sidestream plus mainstream cigarette smoke.

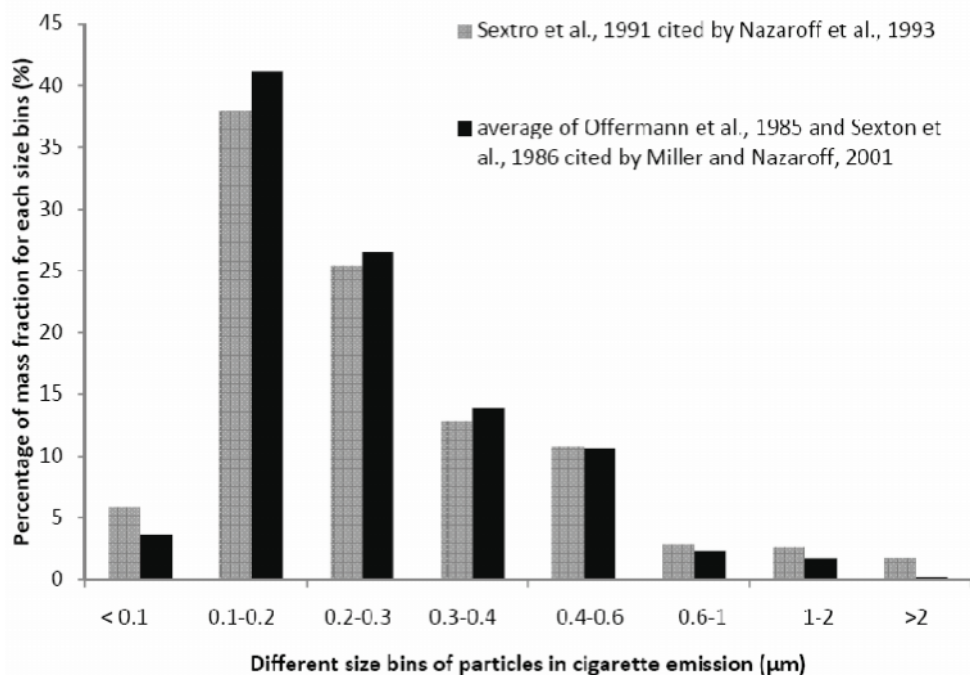


Fig. 2: Percentage of mass fraction for 8 size sections in different reported emission profile for sidestream cigarette smoke.

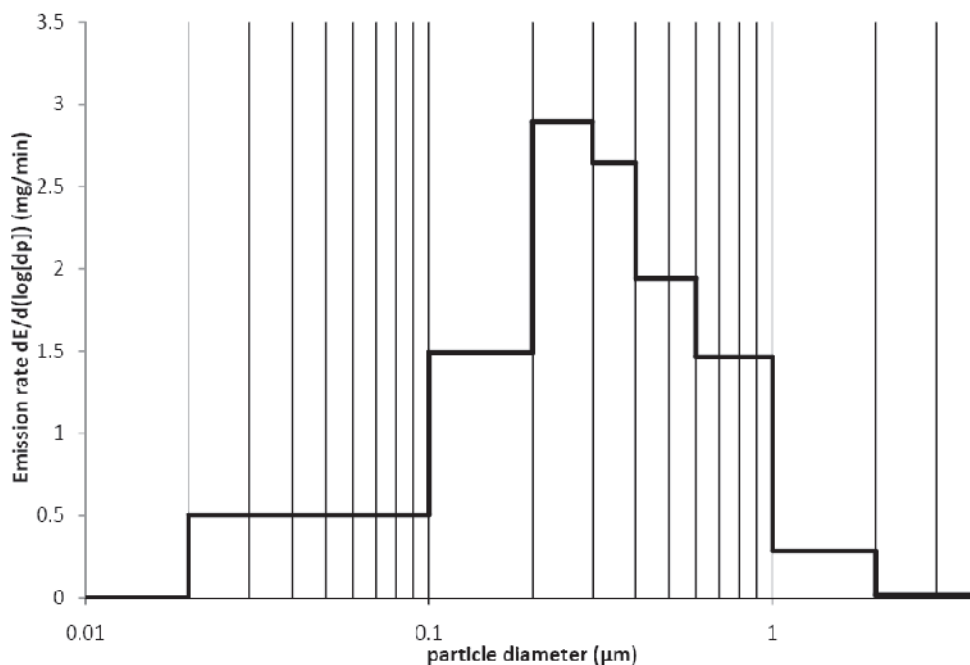


Fig. 3: The assumed emission rate profile of sidestream plus mainstream cigarette smoke as a function of particle diameter.

The assumed emission rate profile of sidestream plus mainstream smoke in this study is shown in Fig. 3. The total mass emission rate is selected based on experimental data of Sextro et al. [11] in which three cigarettes were smoked over a 6 hour period. They reported the mass emission rate of 2.4 mg/min with a mass median diameter (MMD) of 0.48 μm .

The effect of emission source rate on the concentration in each size section is calculated by the following equation:

$$\frac{dC_i}{dt} = S_{E,i} = \frac{E_i}{V} \quad (2)$$

In the above equation, E_i is the emission rate for each size section ($i=1-n$) and the V is the volume of the cabin.

3.2 Ventilation and Filtration

Ventilation system and exchanging the air through the vents and windows of the cars are the main sinks for cigarette pollutants concentration. Selecting the Fresh air mode or the Recirculation mode is the key setting in most of the ventilation system in cars and can influence the amount of air change rate especially in moving cars. When the ventilation system is on Fresh air mode, it mostly exchange the outside air with the air of the cabin through the open vents but when is on the Recirculation mode, it mostly recirculate the inside air. These differences in the operating of the ventilation system lead to higher air change rate in Fresh air mode or when a moving car is on passive ventilation state (the fresh air comes in through the vents without operating of ventilation system).

Nowadays, cabin air filters are mostly installed on cars by many of manufacturer companies. The cabin air filtration duty is to clean the inside air of the cabin and also the external air entering the passenger compartment. The HVAC system of the cars can be equipped by particle filtration (called particle filter) or a combine of particle filtration and the adsorption of gases (called combi filter). Reinhardt and Kobori [12] reported the typical fractional collection efficiency of a combi filter and a particle filter in cars for different particle diameter according to standard test DIN 71460.

The effect of removing rate of the particles caused by ventilation system on the concentration for each size section is calculated by the following equation:

In the above equation, η_i is the filtration efficiency for each size section that affect on the amount of the incoming particles with fresh air and also the rate of removing the particles on recirculation mode. C_{out} is the concentration of particles in fresh air and Q_{Fresh} and Q_{Exit} are the volumetric flow rate of fresh air and exit air that are equal to each other. Q_{Rec} is the volumetric flow rate of recirculating in the recirculation mode of the ventilation system.

3.3 Deposition Model

Deposition of the aerosol particles on the interior surfaces of the cars are characterized by turbulent flow inside the car. The deposition model of Lai and Nazaroff [13] was incorporated in this study for calculating the deposition velocity of each size section from turbulent flow onto smooth surfaces. Considering the surface of seats, dashboard, windows, roof and floor in cars as smooth surface does not lead to noticeable error for RSP particles.

The deposition model of Lai and Nazaroff [13] was applied and extended in many of studies [14-19] and showed good agreement with experimental data. For calculating the deposition velocity of particles on the interior surface of the cars, the complicated geometry of the cabin was simplified to the sum of the vertical, upward horizontal and downward horizontal surfaces. For a relatively high surface-to-volume ratio space like cabin of the cars, the deposition of the particles on interior surfaces is noticeable. The surface-to-volume ratio of a Daewoo Cielo is measured to be 7.33 m^{-1} , that the portions of vertical, upward horizontal and downward horizontal surfaces considered as $1.64V$, $1.64V$ and $4.05V$, respectively.

The friction velocity is the key parameter in the deposition model of Lai and Nazaroff [13] that characterizes the nature and intensity of near-surface turbulent flow. For chambers with low air change rate, the friction velocity is in the order of 0.1 m/s [15] but in the cabin of the cars with high air change rate, the friction velocity is much larger and must be specified. For calculating the friction velocity in each ventilation scenarios of the cars, the experimental data of Ott et al. [20] in cars is incorporated. They reported the deposition rate of the $\text{PM}_{2.5}$ particles for different air change rate in two type's cars (2005 Ford Taurus and 1999 Jeep Cherokee) and found a relationship between the deposition rate (β) and the air change rate of the cabin as $\beta = 1.3 \times \text{ACH}$. Whereas, they reported the overall deposition rate for $\text{PM}_{2.5}$ particles, the calculated deposition velocity for each size section must be converted to overall deposition rate of RSP particles. In first step, the overall deposition rate for each size section is calculated by the approach of Corner and Pendlebury [21]. If the deposition of particles on each surface (vertical, upward horizontal and downward horizontal) act independently in parallel, the overall deposition rate for each size section ($i=1-n$) can be obtained by the following equation [13]:

$$\beta_i = \frac{v_{d_{v,i}} A_v + v_{d_{uh,i}} A_{uh} + v_{d_{dh,i}} A_{dh}}{V} \quad (4)$$

where β_i is the deposition rate for each size section and $v_{d_{v,i}}$, $v_{d_{uh,i}}$ and $v_{d_{dh,i}}$ are the deposition velocity on vertical, upward horizontal and downward horizontal surfaces, respectively. A_v , A_{uh} and A_{dh} and are the total vertical, upward horizontal and downward horizontal surfaces in the cabin of the cars. In second step, the overall deposition rate of RSP particles is calculated based on the portion of each size section in mass emission rate profile of cigarette smoke. The mass emission weighted average of deposition rate is formulated in Eq. 5.

$$\beta = \frac{\sum_{i=1}^s \beta_i E_i}{E_{total}} \quad (5)$$

where β is the overall deposition rate and E_{total} is the total mass emission rate of RSP particles as discussed in section 3.2. In Fig. 2, the results of the deposition model and the experimental data

of Ott et al. [20] are compared to each other. The correlation of friction velocity and air change rate is chosen based on the minimum standard deviation of the model's result with experimental data. By an try and error method, it is found that with the linear correlation of $u^* = 0.2 * ACH + 4.5$, the calculated deposition rate is best fitted on the experiments.

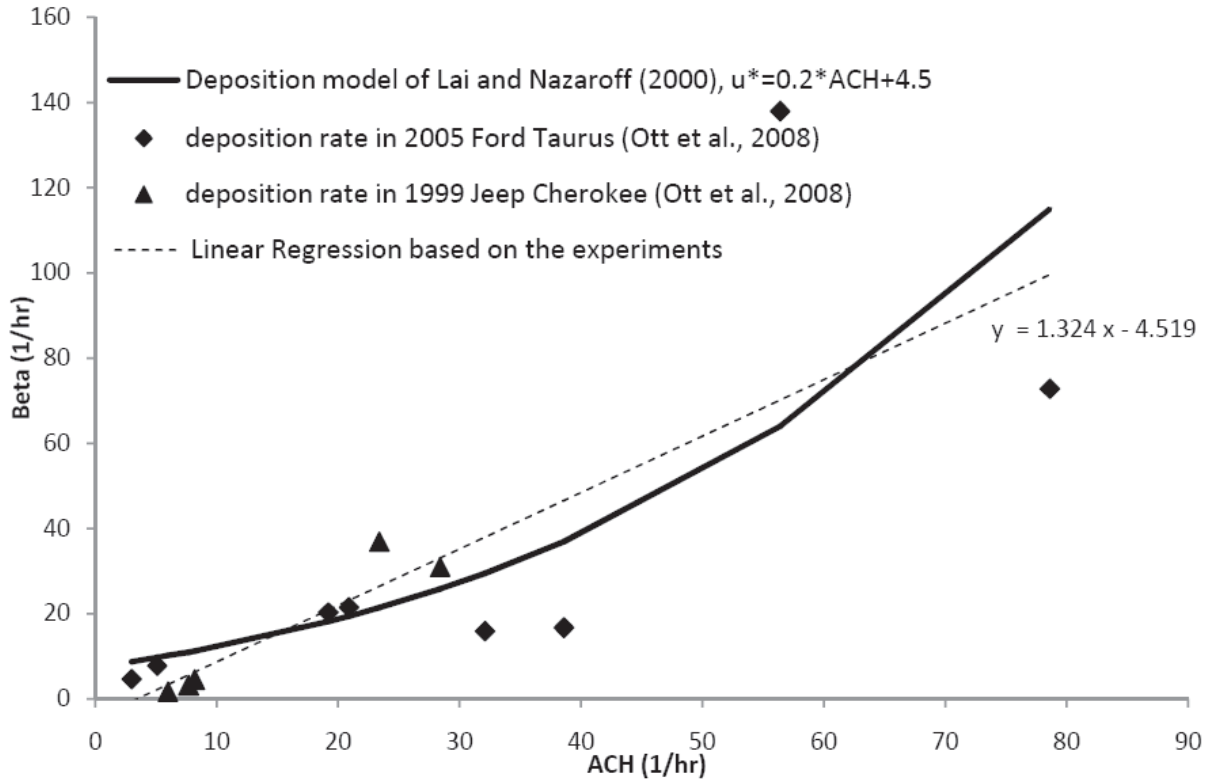


Fig. 4: The prediction of Lai and Nazaroff's deposition model and the measured deposition rate for different air change rate in cars.

The effect of the deposition term on the concentration of each size section can be expressed by the following equations:

$$\frac{dC_i}{dt} = -\frac{C_i}{V} (v_{d_{v,i}} A_v + v_{d_{uh,i}} A_{uh} + v_{d_{dh,i}} A_{dh}) \quad (6)$$

3.4 Coagulation Model

In this study, the coagulation of the particles is calculated based on the same approach that incorporated by Nazaroff and Cass [8]. At first, the collision frequency between two particles is calculated and then, these probabilities is integrated to obtain the generated and removing rate for each size section based on the approach developed by Gelbard and Seinfeld [22].

The coagulation of submicron particles mostly occurs by Brownian motion and other mechanisms like Gravitational coagulation can be ignored for particles generated by cigarette smoke [23]. For calculating the collision frequency of particles through Brownian motion, the interpolation formula of Fuchs [24] is used (Eq. 7).

$$K_B(d_i, d_j) = 2\pi (d_i + d_j)(D_i + D_j) \left[\frac{d_i + d_j}{d_i + d_j + 2(g_i^2 + g_j^2)^{1/2}} + \frac{8(D_i + D_j)}{(d_i + d_j)(\bar{c}_i^2 + \bar{c}_j^2)^{1/2}} \right]^{-1} \quad (7)$$

where, d_i and d_j are the diameter and D_i and D_j are the Brownian diffusion coefficient of the colliding particles. The Brownian diffusion coefficient (D), slip correction factor (C_c), mean particle velocity (\bar{c}) and Fuch's length (g) for each particle can be determined from the following equations [23].

$$C_{c,i} = 1 + \frac{\lambda}{d_i} \left[2.34 + 1.05 \exp\left(-0.39 \frac{d_i}{\lambda}\right) \right] \quad (8)$$

$$D_i = \frac{k_B T_g C_{c,i}}{3\pi\mu_g d_i} \quad (9)$$

$$\bar{c}_i = \left(\frac{48k_B T_g}{\pi^2 \rho_p d_i^3} \right)^{1/2} \quad (10)$$

$$g_i = \frac{\pi\bar{c}_i}{24d_i D_i} \left[\left(d_i + \frac{8D_i}{\pi\bar{c}_i} \right)^3 - \left(d_i^2 + \frac{64D_i^2}{\pi^2\bar{c}_i^2} \right)^{3/2} \right] - d_i \quad (11)$$

In above equations, $K_B = 1.381 \times 10^{-23}$ J/K is Boltzmann's constants and the mean free path is assumed to be $\lambda = 0.0664 \mu\text{m}$ for air at 1 atm and 293 K.

After calculating the collision frequency between each two particles, the sectional coagulation model of Gelbard and Seinfeld [22] is used for determining the source and sink terms in each size section. The mechanism of coagulation acts as sink for smaller particles and as source for larger particles. It is assumed that there are no source terms for particles in the first section and also no sink terms for the last section. The simplified form of this model for a chamber (cabin of the cars) and S sections of the particles are stated in Eq. 12.

where the coefficients of ${}^1a\beta_{pqi}$, ${}^1b\beta_{pqi}$, ${}^2a\beta_{pi}$, ${}^2b\beta_{pi}$, ${}^3\beta_{ii}$, ${}^4\beta_{pi}$ are given in [22]. The adaptive Simpson quadrature [25] is used for evaluating the double integral functions in the correlation of these coefficients. It is noteworthy that these correlations are appropriate for sectional coagulation by considering the mentioned geometric constraint on the mass of the limits in each section ($v_{i+1} \geq 2v_i$, $i=0-n$).

3.5 Solving the Equations

After illustrating the sink and source terms which caused by cigarette emission, ventilation, filtration, deposition and coagulation in cabin of the cars, the total rate of concentration for each size section is calculated by sum of these factors at the same time. The general form of the obtained differential equation for each size section is as Eq. 13.

$$\frac{dC_i}{dt} = S_{E,i} + a_{1,i}C_i + a_{2,i}C_i^2 + a_{3,i}C_{i-1}^2 + \sum_{j=1}^s a_{4,i,j}C_jC_i + \sum_{j=1}^{i-1} a_{5,i,j}C_jC_{i-1} \quad (13)$$

Since the differential equations of concentration in sections ($i=1-n$) are coupled to each other, they must be solved simultaneously. A two-stage implicit Runge-Kutta formula that proposed by Hosea and Shampine [26] is used for solving the 8 non-linear differential equations (8 sections assumed here).

The density of the particles is an important factor and can influence the mechanisms of deposition and coagulation and also the relationship between mass concentration and number concentration of particles. The density of particles depends mainly on their physical properties and chemical composition. Lipowicz [27] measured the cigarette smoke particles density in a millikan cell and recommend the value of $1.12 \pm 0.02 \text{ g-cm}^{-3}$. In this study, the density of the particles is assumed to be 1.1 g-cm^{-3} for all particle sizes.

4 RESULTS

Cabin volume, cigarette emission condition and ventilation scenarios of each reported experiments in the literature (Table 1) are simulated by this model and the results are compared against the experimental data. Whereas, in most of the experimental study in cars, the total $PM_{2.5}$ concentration is measured, the predicted concentration of each size section are summed and presented as total RSP concentration.

The scenario of smoking in cars in each reported experiment [9,20] are simulated and the results of the modeling are compared to experimental data in Fig. 5-6. In most of the cases the modeling can predict the maximum $PM_{2.5}$ concentration and also the decay rate of concentration with a good accuracy.

In Fig. 5 the predicted RSP concentration of Sequential Cigarette Exposure Model (SCEM) that were calculated by Ott et al. [9] were also shown. This model is based on the mass balance equation and predicts a bit higher concentration in comparing to our result. Different emission

rate of cigarette smoke particles that incorporated in each model is the main source of variation in the results.

For illustrating the threats of cigarette smoking on the health of nonsmokers in cars, the mean RSP concentration is compared with EPA health-based standard. The recent EPA health-based PM_{2.5} standard is 35µgm⁻³ for 24 hours. The predicted mean RSP concentration in each simulation for a specified period (during the smoking and decay time until it vanishes) converted into a 24-h incremental exposure (IE₂₄) by the following equation [20].

$$(IE_{24}) = \overline{x_{24h}} = (\text{mean Conc.}) \times \frac{\text{Period of ETS exposure (min)}}{(24 \text{ hr} \cdot \text{day}^{-1})(60 \text{ min} \cdot \text{hr}^{-1})} \quad (14)$$

Using Eq. 14, the 24-h incremental exposures for different car status, windows status and ventilation mode are calculated. It is obtained that the 24-h incremental exposure in stationary cars is high and by smoking two cigarettes (2×30 µgm⁻³=60 µgm⁻³), its value exceeds the EPA's PM_{2.5} limit (35µgm⁻³). Although, the predicted 24-h incremental exposures in moving cars is usually much lower than EPA's limit but when the ventilation mode is on Recirculation, the 24-h incremental exposures is in order of 10 or above. In these scenarios, smoking four cigarettes give IE₂₄=4×10 µgm⁻³=40 µgm⁻³ which is above the EPA's PM_{2.5} limit.

Smokers in the cars could keep some notes in mind to avoid the nonsmokers (especially children) involving in severe conditions in cars. The time needed for each simulated condition to return to initial RSP concentration after smoking a cigarette is also an important parameter. The smokers must prevent the entrance of the nonsmokers until the cigarette pollutant completely vanish (e.g. one hour for stationary cars).

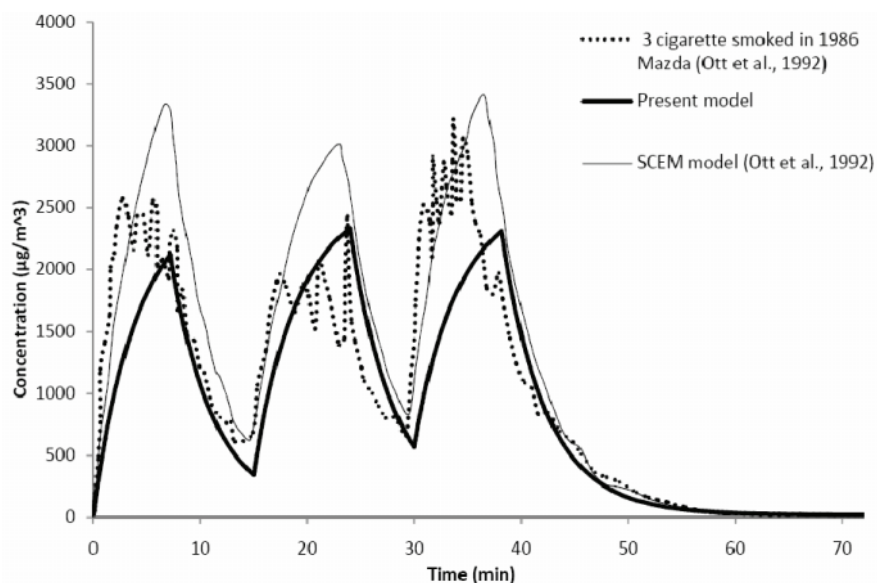


Fig. 5: Comparison of the predicted PM_{2.5} mass concentration in 1986 Mazda with experimental data and Sequential Cigarette Exposure Model (SCEM) result.

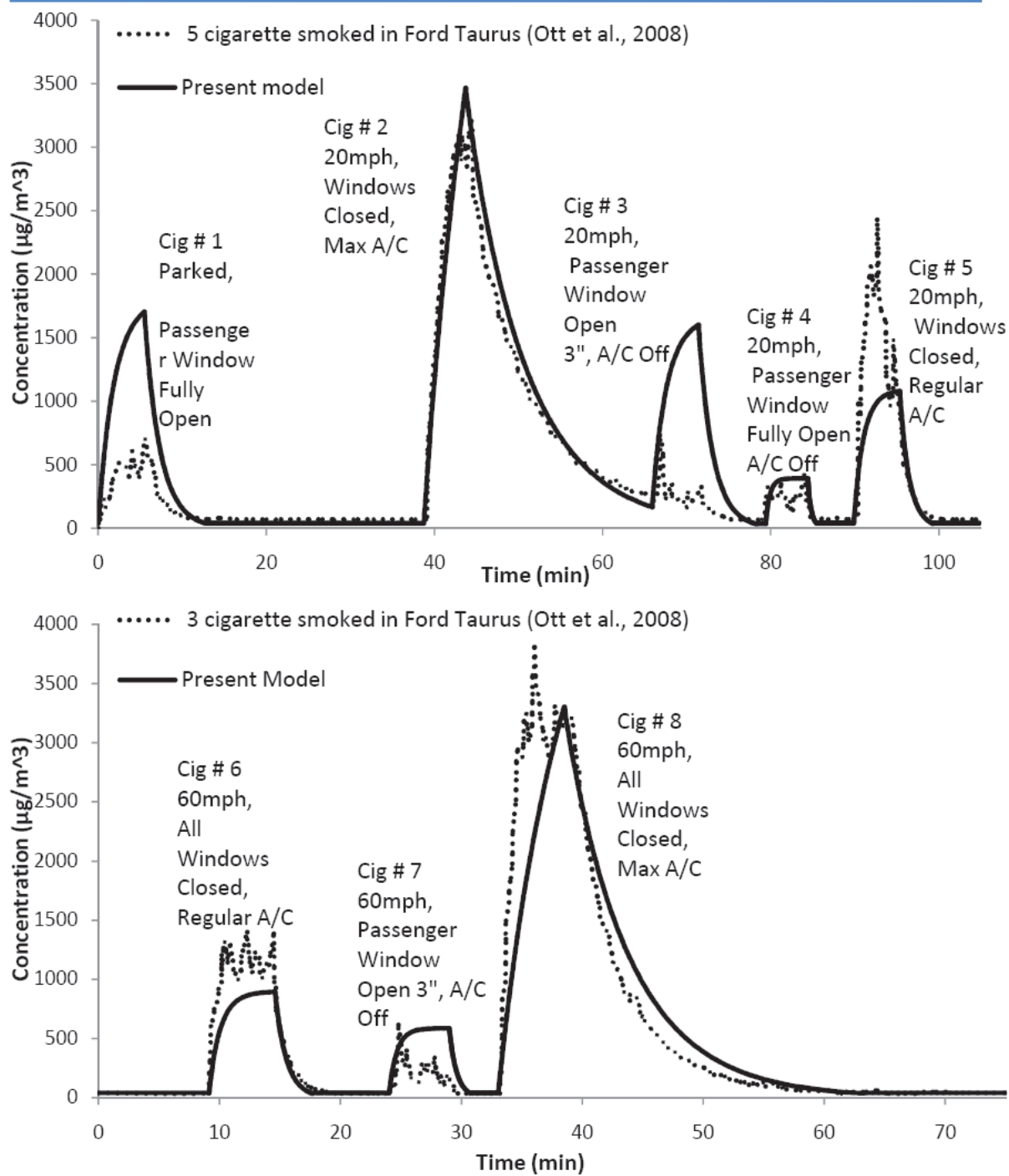


Fig. 6: Comparison of the predicted $\text{PM}_{2.5}$ mass concentration in 2005 Ford Taurus with experimental data at different speed (parked, 20 mph, 60 mph) and ventilation scenarios.



5 CONCLUSIONS

In this study, the dynamics of cigarette smoke particles was simulated in a high air change rate chamber like cars. The mechanism of deposition, coagulation and exchanging the air are taken into account in this modeling and the concentration of respirable particles predicted as a function of time. The scenarios of smoking in the reported experiments in cars are simulated and in the most of the cases the modeling result show good agreement with experimental data. The reported air change rate in the literature on different car status, windows status and ventilation mode are also incorporated in this modeling and the simulation is done for each status. At last, the exposure of nonsmokers in cars compared with EPA health-based $PM_{2.5}$ standard and severe conditions in cars are recognized. The general conclusions reached are as follows:

- In high air change rate chambers like cars, the high turbulence intensity near the walls cause to higher order of deposition velocity in comparison to low air change rate chambers like rooms.
- During the smoking of the cigarette, due to the effect of the deposition on the interior surfaces, the rising rate of particles concentrations decreases by the time.
- In general, by smoking two cigarettes in stationary cars the $PM_{2.5}$ concentration inside the cabin exceeds the EPA health-based $PM_{2.5}$ standard.
- After smoking four cigarettes in moving cars, when ventilation mode is on Recirculation, the 24-h incremental exposure of $PM_{2.5}$ particles exceeds the EPA's limit.
- In general, After smoking a cigarette in stationary and moving cars, the RSP concentration return to initial condition approximately after one hour and one quarter, respectively.

Modern cars mostly are equipped to cabin air filters for protecting the passengers from indoor and outdoor pollutants. More investigation on the efficiency of the filtration in cars and its effect on removing the pollutants is needed. Improving the ventilation system of the cars is being investigated by R&D department of many car manufacturers.

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