

RESEARCH ARTICLE

Particles emitted from indoor combustion sources: size distribution measurement and chemical analysis

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Abstract

This study is primarily focused toward measuring the particle size distribution and chemical analysis of particulate matter that originates from combustion sources typically found in Indian urban homes. Four such sources were selected: cigarette, incense stick, mosquito coil, and dhoop, the latter being actually a thick form of incense stick. Altogether, seven of the most popular brands available in the Indian market were tested. Particle size distribution in the smoke was measured using a scanning mobility particle sizer, using both long and nano forms of differential mobility analyzer (DMA), with readings averaged from four to six runs. The measurable particle size range of the nano DMA was 4.6 nm to 157.8 nm, whereas that of the long DMA was 15.7 nm to 637.8 nm. Therefore, readings obtained from the long and the nano DMA were compared for different brands as well as for different sources. An overlap was seen in the readings in the common range of measurement. The lowest value of peak concentration was seen for one brand of incense stick ($0.9 \times 10^6 \text{ cm}^{-3}$), whereas the highest ($7.1 \times 10^6 \text{ cm}^{-3}$) was seen for the dhoop. Generally, these sources showed a peak between 140 and 170 nm; however, 2 incense stick brands showed peaks at 79 nm and 89 nm. The dhoop showed results much different from the rest of the sources, with a mode at around 240 nm. Chemical analysis in terms of three heavy metals (cadmium, zinc, and lead) was performed using graphite tube atomizer and flame-atomic absorption spectrophotometer. Calculations were made to assess the expected cancer and noncancer risks, using published toxicity potentials for these three heavy metals. Our calculations revealed that all the sources showed lead concentrations much below the American Conference of Governmental Industrial Hygienists (ACGIH) threshold limit value (TLV) level. One of the two mosquito coil brands (M.) showed cadmium concentrations two times higher than the California Environmental Protection Agency (Cal EPA) reference exposure level (REL). The latter also showed the highest carcinogenic risks of 350 people per million population. The amount of zinc obtained from the sources, however, was found to be quite below the standard limits, implying no risk in terms of zinc.

Keywords: Metal toxicity; indoor combustion sources; size distribution of smoke; smoke inhalation risk

Introduction

Indoor air quality has a very strong influence on well-being and productivity, with indoor pollutants increasing the risks for a wide array of diseases. A study by Jenkins et al. (1992) showed that people spend 87% of their time indoors, with only 6% outdoors, and 7% in transit. It also poses a threat to materials, for example, paintings and electronic equipment (Nazaroff & Cohen, 2004). Starting in the middle of 1970s, it was found out that the indoor concentrations of several substances were appreciably higher than that in outdoors. These substances were identified to be combustion by-products, volatile organic compounds, radon, and bioaerosols. This work primarily focuses on indoor combustion by-products found in a typical urban Indian household.

Smoke from indoor combustion sources has been one of the oldest indoor air problems worldwide. Biomass fuels like wood, animal dung cakes, charcoal, and coal can give rise to high concentrations of particulate matter (Venkataraman et al., 2001). It is the eighth leading cause in reducing human life span, with its effect being felt strongly among women and young children (Samet & Spengler, 2003). Most of the studies done in the past within Indian scenario have dealt with the problem of indoor biomass combustion within rural households (Venkataraman et al., 2001).

One of the main reasons for smoke being a major indoor problem is its wellspring of heavy metals. Heavy metals affect the immune system and the central nervous system, and also cause depression (State of California Air Resources

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Board, 2005). Leading among these are lead, cadmium, and zinc. Heavy metals in smoke are connected with heart-rate variability (Magari et al., 2002) and an increase in the number of respiration related emergency cases (Clairborm et al., 2002). Lead, a very toxic element, causes brain and kidney damage for short exposure times and affects the central nervous system and vitamin D metabolism in the long run (U.S. EPA, 2000). Knowledge about its carcinogenic effects still remains scant, thereby leading it to be classified as a Group B2 (probable) human carcinogen. For short times of exposure, cadmium causes pulmonary irritation in the lungs. Chronic effects include kidney and lung disease, including the formation of kidney stones (U.S. EPA, 1999). Zinc is present as a gaseous pollutant primarily as zinc chloride or zinc oxide. Inhaled zinc gives rise to metal fume fever, whose symptoms are a metallic taste in the mouth accompanied by a dryness and irritation in the throat, followed by flue-like symptoms, chills, fever, profuse sweating, headache, and weakness (Choudhury et al., 2005).

Morselli et al. (2007) have assessed and compared the environmental performances of a regional incinerator network. They have emphasized that special attention must be paid to Cd and As, which are toxic even in low quantities. Valberg et al. (1996) have enumerated methods for toxicity modeling and risk assessment for heavy metals in air. Hogervorst et al. (2007) threw light on house dust as a possible source of cadmium and lead in indoor environments. Damek-Poprawa and Sawicka-Kapusta (2002) found that even relatively low concentrations of lead and cadmium caused damage to the kidney, liver and testis of yellow necked mice in Poland.

The common indoor combustion sources seen in typical urban Indian homes are cigarettes, incense, and mosquito coils. A fair number of particle size distribution studies of cigarette smoke and its subsequent temporal evolution have been reported in the literature mostly for the developed countries (Morawska et al., 1999; Okada & Matsunuma, 1974; Ishizu & Okada, 1978; Ueno & Peters, 1986; Ingebretsen, 1986), and there is a great degree of mismatch between them, which could primarily be attributed to the measurement techniques employed.

Okada and Matsunuma (1974) measured the particle size distribution and concentration of a cigarette smoke by a light scattering method. The geometric mean diameters were measured to be 180 nm and 100 nm for the mainstream and sidestream smoke, with the number concentration of the mainstream smoke being $3 \times 10^{10} \text{ cm}^{-3}$. They also experimented with five different brands of cigarette. They concluded that the number concentration in each of these cases was $\sim 10^{10} \text{ cm}^{-3}$, and the value of the mode radius was between 0.135 to 0.16 μm . Ingebretsen (1986) has studied the evolution of the number distribution of the mainstream cigarette smoke aerosol, concluding that the number-averaged diameter reached a peak, decreased, and then increased again with time. The particle size was seen to correlate with the changing flow rate. Ishizu et al. (1977) have analyzed the changes in particle size and number density of cigarette smoke through the column of a cigarette,

by varying puff volume with time, using a light-scattering photometer for the measurements. They concluded that the geometric mean diameter increases and the number density decreases with time or decreasing puff volume. These conclusions pointed to the fact that as the residence time of the cigarette aerosol became higher, the geometric mean diameter increased and the number concentration decayed. Morawska et al. (1999) investigated the deposition of environmental tobacco smoke in the human respiratory tract. They conducted measurements with 15 nonsmoking and 3 smoking volunteers. The smoke these subjects inhaled had a count median diameter of 200 nm and a geometric standard deviation of 1.7. The authors used a scanning mobility particle sizer to measure the particle size distribution in the inhaled and exhaled air. Ueno and Peters (1986) analyzed the generation rate and size distribution of sidestream smoke using two instruments: an electrical mobility analyzer and a cascade impactor. They found that the number mean diameter and mass median diameter of the smoke were around 0.1 μm and 0.16 μm , respectively, and that the distribution was broader compared to the mainstream smoke, indicating thereby a greater level of heterogeneity.

The goal of this study was to measure the particle size distribution in smoke emitted by indoor combustion sources commonly found in Indian homes. India, being a tropical country, suffers largely from mosquito-borne diseases and thus the majority of the households especially in the urban area rely on either mosquito coils or net. Mosquito coils are generally used overnight. However, some affluent families do employ expensive liquid mosquito repellants. Incense and cigarettes are also quite extensively seen in urban homes. Only recently, cigarette smoking at public places including pubs has been banned throughout India. This has been done to control passive smoking in the larger interest of the public. The focus was on indoor combustion sources typically found in urban environments. To the best of our knowledge, no study on the particle size distribution and toxicity has been done so far on indoor smoke sources in India, other than the indoor biomass sources, especially in rural households. While measuring, the particles were sampled as they were emitted from the source, without any dilution. Finally, the heavy metal content in these samples was also measured, and a cancer and non-cancer risk assessment of each source from each metal was performed.

Methodology

Experimental setup

The particulate size and number distribution measurement

In total, four different combustion sources and seven different brands were selected for experiment. These were cigarette (C_1, C_2) incense stick (I_1, I_2), mosquito coil (M_1, M_2), and dhoop (D_1). The dhoop is a type of thick incense stick. The

cigarettes were filtered. No modification was done on any of the sources, and the smoke measured was directly from the source.

The experimental setup used is depicted in Figure 1. The combustion source was clamped to a burette stand. A funnel was held over the combustion source to act like a mini-hood, in order to completely capture the smoke and prevent it from spreading out and dispersing. At the funnel, the flow was split into two parts. One part went to the scanning mobility particle sizer (SMPS) for sampling, and the rest went to the vacuum pump. The vacuum pump was used to dump the excess smoke. The sample flow rate for the SMPS was maintained at 0.3 lpm (L/min). It was measured by a thermal mass flow meter, supplied by TSI. The dotted suction line beneath the combustion source means that it was used only to provide suction to a cigarette to burn effectively, and was redundant in cases of other combustion sources. The pump used was a diaphragm vacuum pump. The conical flask shown was used as a dampener to minimize any oscillations in the flow rate.

The instrument used for particle size measurement was a scanning mobility particle sizer (TSI, model 3093). The electrostatic classifier (model 3080) was used with long DMA (model 3081) and nano DMA (model 3085). A scan time of 90 s followed by a retrace time of 30 s was used for both the DMAs. This was found to be optimum to measure accurately the size distribution of sampled smoke. The particle size range measured on the nano DMA was 4.61 nm to 156.8 nm, whereas it was 15.7 nm to 637.8 nm for the long DMA, suggesting an overlap between the two measurement ranges. Multiple charge correction was not used, since its presence would distort the size distribution from the actual. Only one scan was done per sample. A sample here means a reading taken every 2 min. Six samples were taken for each source, and the readings obtained were averaged by neglecting the first sample, since it was considered that the first reading contained some signature remnant of the previous source.

The sampling of indoor smoke

Sampling smoke from each source was done using a cascade impactor loaned from Bhabha Atomic Research Centre (BARC), Mumbai. This impactor was designed by Singh et al. (2005) and can separate aerosol particles into 7 size bins from 0.53 μm to 10 μm ; these sizes are the cut points of 50% efficiency pertaining to a given Stokes number (Friedlander, 2000; Hinds, 2001). A schematic of the experimental setup is shown in Figure 2. The stage cutoff diameters were 10 μm , 7.7 μm , 5.5 μm , 3.5 μm , 2.5 μm , 0.98 μm , and 0.53 μm , respectively. Each source was allowed to burn for 1 h, and a vacuum pump was connected to the impactor to draw the smoke sample through the impactor at a flow rate of 45 lpm as recommended by the impactor design (Singh et al., 2005). A piece of quartz filter paper (high porosity, QMA TSP/PM10 grade, Whatman, Inc., USA) was placed in the final filter of the impactor to collect the sample of submicrometer particles. The 0.53 μm impaction stage was not used since we wanted to collect all submicrometer particles to be collected onto a single final filter. Removal of the 0.53- μm impaction stage led to the reduced overall pressure drop through the cascade impactor. Note that the cascade impactor was purely used to collect only the submicrometer particles to facilitate further chemical analysis for key metals. Here the cascade impactor was not used to determine the size distribution of the sampled smoke.

The measurement of chemical composition: instruments used

Detection of metals in the samples was accomplished by means of two types of atomic absorption spectrometry (AAS), namely, the graphite tube atomizer (GTA; Varian Spectra AA-20 GTA-96, Australia) and the flame emission spectrophotometer (Varian Spectra AA-220 FS, Australia). The flame emission spectrophotometer was used to measure the high Zn content of the filters. The concentrations of Cd and Pb were low; hence the GTA was used for higher accuracy. Both the GTA and the AAS were calibrated using the analytical (working) curve technique (Willard et al., 1986),

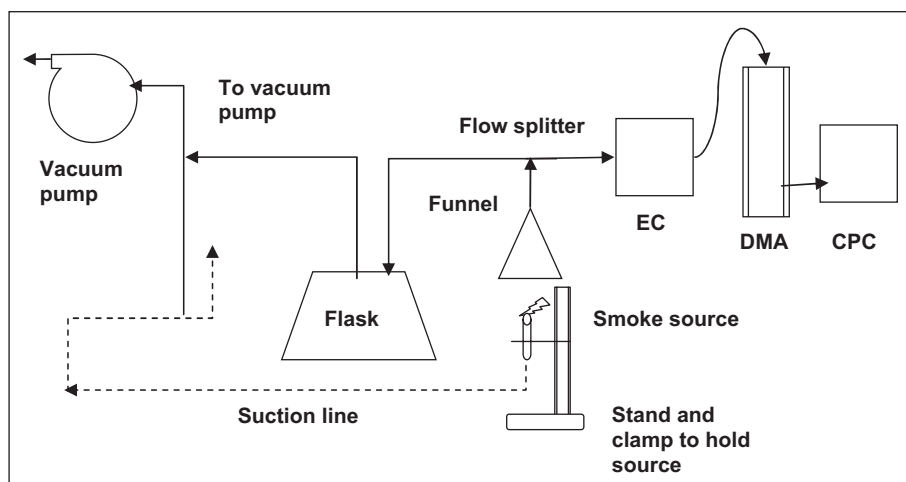


Figure 1. Set up for the experiment to measure particle size distribution by the SMPS. Abbreviations used: EC- Electrostatic Classifier, DMA - Differential Mobility Analyzer, CPC - Condensation Particle Counter.

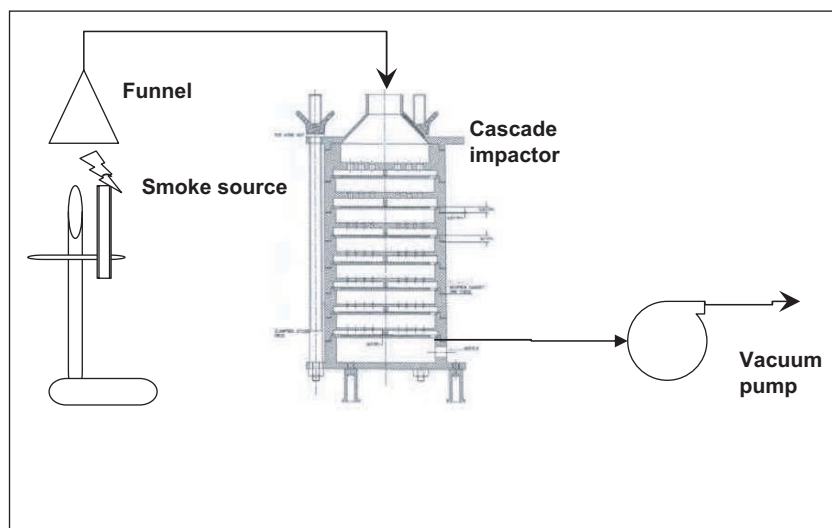


Figure 2. A schematic of the set-up used for sampling of smoke for subsequent chemical analysis.

and the standards for the same were prepared using the U.S. EPA Compendium Method IO-3.2.

Calculations

Relative amounts of metals present in each sample

This gives the difference between the metal concentration in the sample and the metal concentration in the blank. It is calculated by the following equation (Chinnam, 2005):

$$C_i = (F_s - F_b) \quad (1)$$

where C_i is the concentration of metal in sample ($\mu\text{g L}^{-1}$), F_s is the concentration as obtained from AAS ($\mu\text{g l}^{-1}$), and F_b is the concentration in blank, as obtained from AAS, ($\mu\text{g L}^{-1}$).

Metal content per source

This method used to calculate metal content per unit source is drawn on the lines of the one used in the calculation of exposure relevant emission factors in Singer et al. (2003). The method used is drawn on the calculation delineated in Chinnam (2005).

We have

$$C = C_i V / N_s \quad (2)$$

where C_i is the concentration of metal in sample in micrograms per liter, from Eq. (1), V is the makeup volume, 0.1 L, and N_s is the number of source units burnt.

Estimation of cancer and noncancer risks

The metal mass as obtained in filter paper (calculated by multiplying the metal concentration with the makeup volume) was normalized to the volume of a typical room ($10\text{ft} \times 12\text{ft} \times 10\text{ft}$, approximately 35m^3), in order to obtain the metal concentration in indoor air.

$$C = C_i V / V_r \quad (3)$$

where V_r is the volume of the room, as delineated earlier.

The values of metal concentrations so obtained were then compared with the standard values reported by various regulatory agencies. A carcinogenicity risk assessment for cadmium was also conducted. This was done by dividing the metal concentrations in smoke by that corresponding to causing a risk of 1 in a million. Calculations for personal exposure and Lifetime Cancer Risk (LCR) are shown in the appendix, performed using the method published in Kammen and Hassenzahl (2001). Although this study is providing very little data to actually conduct any major risk assessment, the preceding exercise excited us to get some preliminary numbers. Based on these data, more extensive study can be carried out in future.

Possible sources of error

Errors can crop up in all the three components of the experiment, namely, (1) chemical composition measurement, (2) sampling, and (3) size measurement

A newly formed pure metal atom is inherently unstable. It tends to combine with other elements to reach a minimum energy configuration. Consequently, it has a tendency to combine with other elements present in the flame. These metal compounds so formed may also absorb and emit radiation. These emitted radiations will interfere with those emitted by the pure metal atoms, and thereby give rise to errors in the measurement by the AAS.

The graphite tube atomizer can produce metal atoms with more efficiency than the flame AAS, primarily because there are none of the reactions just described that tend to consume the freshly formed pure metal atoms. However, they can maintain the high concentration for a relatively shorter time as compared to the flame AAS. This is because the GTA can maintain the absorption signal only when the sample is introduced, whereas the flame AAS can maintain it as long as the solution is being atomized. Therefore, any change in the atomization rate in the GTA during the relatively available short time for measurement of radiation may possibly give rise to serious analytical errors. The readings

obtained for our samples fell on the curve formed by the standard solutions used for calibration, thereby validating our results.

Problems in sampling include depositional losses to the wall of the impactor due to diffusion (Hinds, 2001). Other losses include thermophoretic and electrostatic wall effects (Baron & Willeke, 2001). These losses manifest themselves more for small particles, and can be offset by using a long sample time, ~ 1 h.

In the measurement of size for submicrometer particles, as the aerosol is led to the charge neutralizer and then to the DMA, diffusion, electrostatic attraction, and gravitational settling may contribute to losses, especially since the inlet is made of metal, a conductive material. So these losses were definitely there in the setup, in the funnel and in the pipes that led to the SMPS inlet. Also there could have been losses brought about by the ration of ambient to sampling velocity. Sampling of too few particles may result in a displayed size distribution not characteristic of the true one (Baron & Willeke, 2001, pp. 125–127). However, these errors would not affect the physical trends of the aerosol size distributions that were measured.

Results and discussion

The particle size distribution: an overall picture comprising of both DMAs

Figures 3 to 6 show the particle size distributions of the smoke emitted from the various combustion sources, as per the different brands. These readings are the averages of four to five samples, as the size distributions reach a steady state quickly. The measurements of the nano and the long DMA are superimposed on each other, in order to give the entire size range of particles emitted from these sources. The error bars indicate that the readings are averaged out from four to five samples, with the bars themselves showing the standard deviation. The y axis shows the number of particles present per unit volume of air sampled, and the x axis the mobility diameter.

In Figure 3a, we see the size and number distribution for particles emitted from the cigarette of brand C_1 . For both the nano and the long DMA, we see a minimum at around 22 nm. It is also seen that the readings overlap in the region common to the measurement range of both the instruments. A difference in readings obtained by the two DMAs could be attributed to the different residence times of the aerosol streams in the two different DMAs, primarily since the volumetric holdups in them are unequal. A maximum is seen at around 160 nm. Similar trends are seen for the cigarette of brand C_2 in Figure 3b, with the only marked difference being that the mode is seen at 150 nm. Similar data were recorded by Okada and Matsunuma (1974), who also reported the mode to be at around 180 nm for the main smoke stream, but the numerical value of their peak concentration was around $3 \times 10^{10} \text{ cm}^{-3}$. This difference could be due to the difference in brands, and thereby composition of the cigarettes.

The particle size distribution for the mosquito coil M_1 is shown in Figure 4a. Here, the nano DMA shows an initial minimum at around 22 nm, whereas the long DMA shows the minimum at about 27 nm. The readings overlap in the common range of measurement. A peak is seen at 150 nm and a linear, downward trend thereafter. Similar trends are seen for the mosquito coil M_2 in Figure 4b, with the only difference being that the nano DMA shows an initial minimum. Figure 5a shows the size distribution of smoke emitted by incense stick of brand I_1 . Here, the initial minimum from both the DMAs is at around 19 nm. An interesting fact is that the mode is seen at around 90 nm, which is little more than about half that of the earlier sources. However, the value of the number density at the mode is of the same order of magnitude, i.e., 10^6 cm^{-3} , the sources described earlier. These results again show up in Figure 5b for the incense stick of brand I_2 , the mode being seen at 79 nm. In Figure 6, we see the particle size distribution for smoke from a dhoop of brand D_1 . Here, the nano DMA shows an initial minimum at 22 nm, while the long DMA shows one at 31 nm. The peak is seen at around 240 nm, a much higher value as compared to other sources. Also, the size distribution of particles from dhoop (D_1) assumes

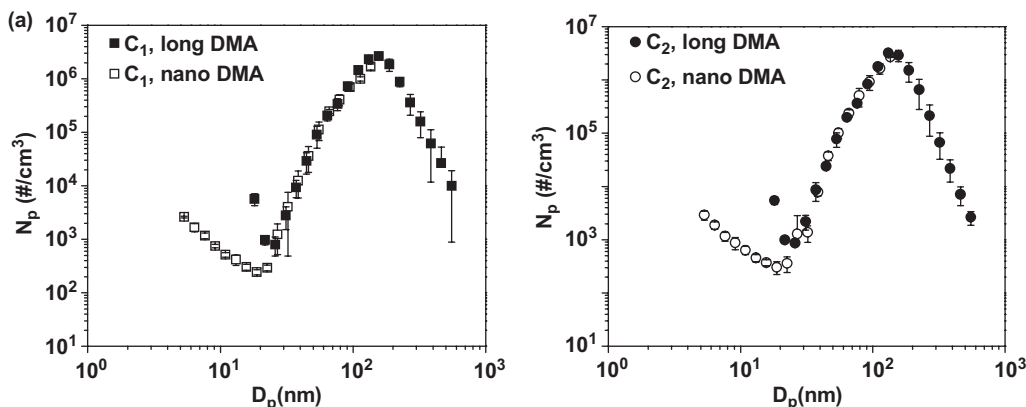


Figure 3. Particle size distribution of smoke from cigarette (a) C_1 (b) C_2 , as measured by SMPS. Error bars represent variability in the data.

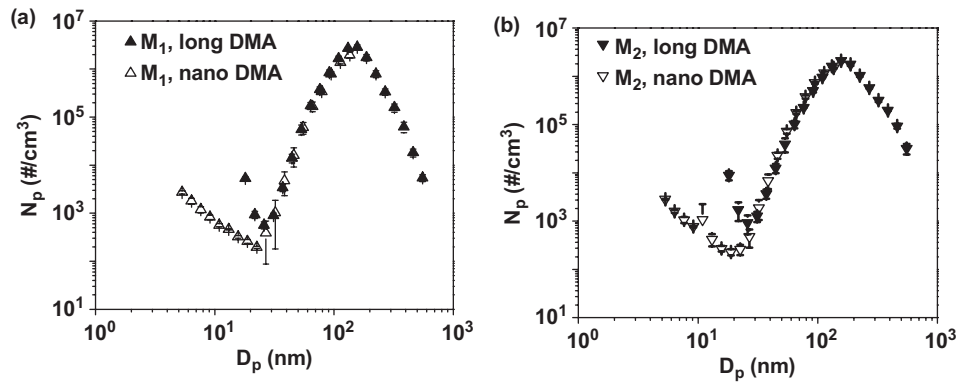


Figure 4. Particle size distribution of smoke emitted from mosquito coil of (a) M_1 and (b) M_2 , as measured by SMPS.

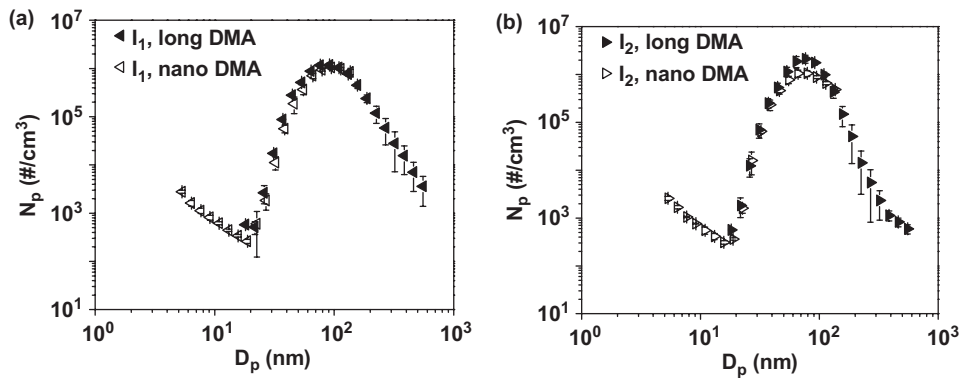


Figure 5. Particle size distribution of smoke emitted from incense stick of (a) I_1 and (b) I_2 , as measured by SMPS.

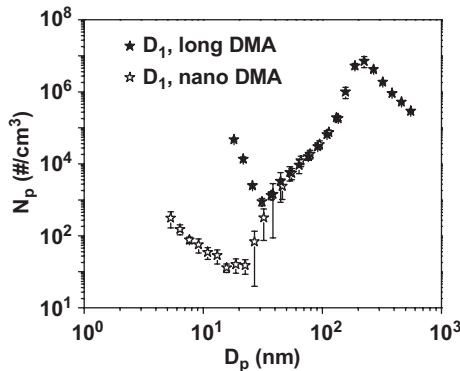


Figure 6. Particle size distribution of smoke emitted by dhoop D_1 , as measured by SMPS.

a markedly different shape as compared to the fairly bell-shaped curve of the other sources. The reason for dhoop showing a different size distribution and significantly higher mode than all other six smoke sources is due to the inherently denser smoke stream from it, as compared to the other smoke sources. This higher number density gives rise to an increased coagulation, implying larger particle size and thereby a higher value of the mode. The values of $t_{1/2}$ are also calculated and presented later in Table 1.

Evolution of the aerosol size distribution

Figures 7 to 10 show the time evolution of the aerosol for the different sources, the readings being recorded at an interval of 2 min. The plot range for the nano DMA was chosen as 4–15 nm, and that for the long was 15 nm onward, so as to prevent overlap. Plots from just one brand for each source are presented here, as similar plots were obtained even by burning different brands of a given source. The first sample, i.e., the one coming after 2 min, was not considered since there is always some residual volume of the previous sample inside the DMA, which caused some starting noise.

Figure 7 shows the time evolution of smoke from cigarette of brand C_1 . It is seen that for the long DMA readings all the three graphs are coincident for sizes up to the mode. However, they differ in terms of the number of particles larger than the mode diameter. Okada and Matsunuma (1974) reported that the distribution remains qualitatively the same but changes quantitatively with time. Also, in 1999 using the numerical model Robinson and Yu claimed that the size distribution progressed to a flatter and wider one in 2 s. Our results are in better agreement with the findings of Keith and Derrick (1960), who found that the size distribution remained unchanged with butt length and therefore with respect to time. The first graph, in all the smoke sources, shows some difference with respect to those seen at later times, which could be attributed to starting noise. For the

Table 1. Source, mode diameter, burn-time, characteristic half-time and doubling time and corresponding number density of the size distribution for the different sources used in this study.

Source	Brand	Stick Burnt (number)	Modal Diameter (nm)	$N_{p,mode} \times 10^{-6}$ (#cm ⁻³)	Burn-time (mins)	$t_{1/2}^1$ (mins)	t_2^2 (hrs)
Cigarette	C ₁	10	157	2.7	8	11	1.3
	C ₂	10	136	2.7		10.6	
Incense	I ₁	15	89	0.9	10	22	3
	I ₂	15	79	2.1		9.8	
Mosquito	M ₁	¼	157	2.8	60	10	1.3
Coils	M ₂	¼	157	2.1		13.4	1.3
Dhoop	D ₁	1	238	7.1	60	4.6	0.5

¹The $t_{1/2}$ value corresponds to the time required for the peak concentration to decay to one-half its initial value.

²The t_2 value is the time required for the mode diameter to double because of coagulation. These characteristic time calculations were performed using the values of coagulation kernels given in Appendix A11, Hinds (1999).

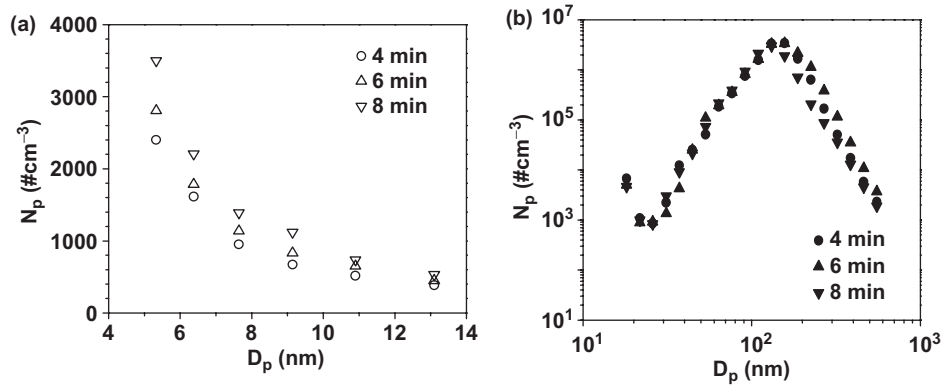


Figure 7. Time evolution of particle size distribution from source C₂ (a) on nano DMA, (b) on long DMA.

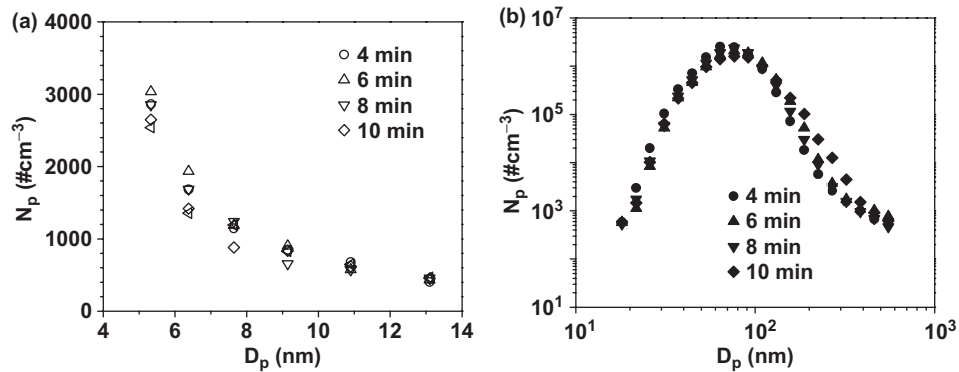


Figure 8. Time evolution of particle size distribution from source I₂, (a) on nano DMA (b) on long DMA.

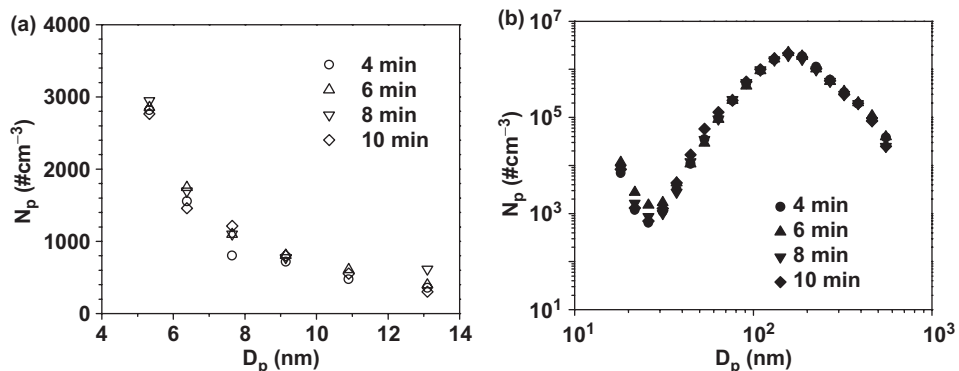


Figure 9. Time evolution of particle size distribution from source M₂ (a) on nano DMA (b) on long DMA.

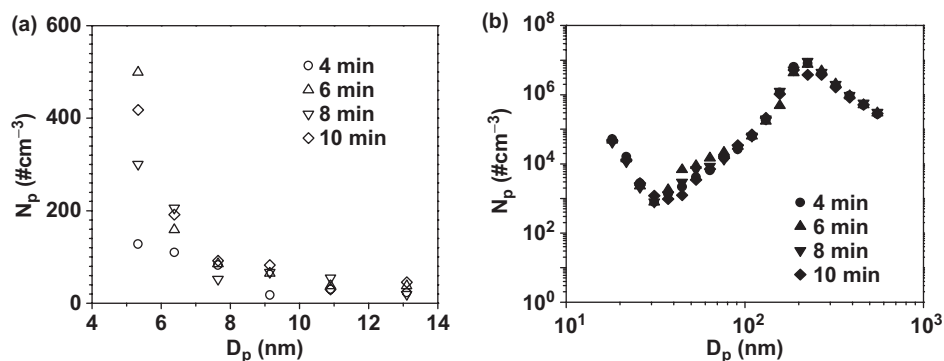


Figure 10. Time evolution of particle size distribution from source D_2 (a) on nano DMA (b) on long DMA.

readings on the nano DMA, we see that the size distributions in the range of 8–15 nm reach a steady state by falling on the same curve, but the same cannot be said about those from 4–8 nm, as some marked differences remain. The best match is seen in the size range of 10–15 nm. Similar results are seen in the plots for other sources. Also, the long DMA curves for the mosquito coils and dhoop agree much more closely than those for other sources. As for the other sources, namely, the cigarettes and incense, this agreement is seen up to the mode, and some deviation is seen thereafter. As seen in Figures 3 and 5, the standard deviations seen in the distributions beyond the mode are quite significant, and this variability, as seen in the time evolution, could well be the cause for it. Also, the number distributions of dhoop, as seen on the nano DMA, are one order of magnitude less than those seen for other sources.

Table 1 shows the source, mode (peak), and its corresponding numerical value, as seen in Figures 3 to 6. It can be seen that the maximum value of the mode occurs for the dhoop, whereas the minimum value is seen for one brand incense sticks. Also, the mode for dhoop comprises of particles of size around 240 nm. This larger size could perhaps be due to enhanced coagulation in the inherently dense dhoop aerosol. We know that the removal efficiency curve for particles (Hinds, 2001; Friedlander, 2000; Willeke & Baron, 2001) shows a minimum between 100 and 400 nm. Particles smaller than 100 nm get removed from a given space by diffusion, while those larger than 1000 nm get removed efficiently by sedimentation. There is thus an intermediate range in which no removal mechanism works efficiently. This range is called the accumulation mode, and falls in a window of 100 to 400 nm. These particles are difficult to remove and persist in the environment. Consequently, these particles will have a higher tendency to persist in indoor environment and thereby be available for uptake.

Table 1 also gives an estimate of the time required for a source to completely burn. It can be inferred that since mosquito coils and dhoop have the highest burn times, they will have a tendency to persist in the indoor environment for a longer time. The mosquito repellent coils are burnt mostly during the evening and night (least human activity). This gives the possibility of a longer exposure time for an individual, and thereby arises the possibility of posing a greater

risk. Also calculated were the values of $t_{1/2}$ and t_2 (Table 1), which are the characteristic times for the mode number concentration to reduce to half and the mode diameter to double, respectively. It is seen that the dhoop has the shortest characteristic times for both the cases, with the longest being for the case of incense stick. So the incense particles will have a greater tendency to persist in the indoor environment. And even when their mode diameter doubles, they will still stay around 150 nm. Morawska et al. (1999) showed in their study that particles of diameters around this value readily deposit in the lungs. It can be concluded that the incense stick smoke will have a greater tendency to deposit in the lungs, and cause injury.

Chemical analysis

Table 2 shows, for each source, the raw data pertaining to the mass obtained on filter paper. The data obtained in Table 1 are then normalized per unit source using Eq. (2) and Table 2. The results are shown in Table 3.

As seen from Table 3, the mosquito coil M_2 contains the highest amount of lead among all the sources. The smallest amount is found in the cigarettes. The Cd levels too were high in M_2 , with the lowest amount being seen in the dhoop. The levels for Zn are higher than those for Cd and Pb, with dhoop being the highest source. Also, as seen from Table 3, the concentrations of Cd in the smoke are the lowest, from half a nanogram to a few nanograms, with the highest levels being those of Zn.

Calculation of cancer and noncancer risk

Table 4 shows the concentrations of metals in smoke, normalized to volume of a room, as calculated by Eq. (3). It is from the data in this table that the toxicity comparisons for the various metals are done, by comparing them with various standards, given in the next subsection.

Comparisons with standard data

Table 5 shows the comparisons for cadmium, lead, and zinc, comparing the calculated values vis-à-vis the standards of various regulatory organizations.

The cadmium values here are compared against the Cal EPA reference exposure level (REL), since that is the lowest dose value among all the values displayed, and thereby

Table 2. Metal Content in Samples. All values are in $\mu\text{g l}^{-1}$.

Metal Content Sample Type		Pb			Cd			Zn ³
		A ₁ ⁴	A ₂	A _{avg}	A ₁	A ₂	A _{avg}	A _{avg}
Cigarette	C ₁	9.45	14.49	11.97	0.99	0.88	0.94	230
	C ₂	4.53	0.16	2.35	1.08	0.53	0.81	166
Incense	I ₁	103.35	95.74	99.55	4.96	5.2	5.1	363
	I ₂	56.95	73.15	65.05	1.24	1.86	1.55	1201
Mosquito Coils	M ₁	108.41	104.83	106.62	1.64	1.41	1.53	127
	M ₂	239.83	229.25	234.54	71.86	74.81	73.34	264
Dhoop	D ₁	180.55	175.08	177.82	0.15	0.22	0.18	628.5

³The software coupled to the Flame-AAS directly calculates averages without displaying individual results, so for Zn only average of two readings are shown.

⁴A stands for actual value, corrected against the blank. The subscripts 1 & 2 correspond to injections 1 & 2 to the AAS.

Table 3. Metal content per unit source. All units are in $\mu\text{g}/\text{source}$.

	Cigarette		Incense		Mosquito Coil		Dhoop
	C ₁	C ₂	I ₁	I ₂	M ₁	M ₂	
Pb	0.12	0.024	0.66	0.43	42.65	93.82	17.78
Cd	0.0094	0.008	0.034	0.01	0.61	29.34	0.018
Zn	2.30	1.66	2.42	8.0	50.8	105.6	62.85

Table 4. Metal concentration in smoke in a typical room (volume 35 m³): contribution of sources of various types. All units are in $\mu\text{g m}^{-3}$.

	Cigarette		Incense		Mosquito Coil		Dhoop
	C ₁	C ₂	I ₁	I ₂	M ₁	M ₂	
Pb	0.034	0.0067	0.28	0.18	0.3	0.67	0.51
Cd	2.7×10^{-3}	2.3×10^{-3}	0.015	4.4×10^{-3}	4.4×10^{-3}	0.21	5.25×10^{-4}
Zn	0.66	0.47	1.04	3.43	0.36	0.75	1.8

would involve a larger factor of safety. It is observed that except for one of the mosquito coils (M₂), all the other sources fall well below the prescribed value. It can thereby be inferred that this mosquito coil M₂ has the highest chance of causing damage, since the concentration of cadmium in the smoke emitted by it is about two times higher than the Cal EPA REL. The comparisons for lead are done with respect to American Conference of Governmental Industrial Hygienists (ACGIH) threshold limit value (TLV), for reasons the same as those given in the assessment for cadmium. It is seen that all of the sources have lead content way below the ACGIH TLV level. Similar trends are seen for zinc as for lead. In fact, zinc levels found in all the analyzed smoke samples were far below the CAL EPA REL values.

Carcinogenic risks

As per the U.S. Environmental Protection Agency findings (U.S. EPA, 1999), a concentration of Cd of $6 \times 10^{-4} \mu\text{g m}^{-3}$ is likely to cause cancer in 1 person out of a population of 1 million people. Dividing the concentrations which pertain to unit volume in a room by this value, one can ascertain how many people are prone to this risk. The results are tabulated as follows in Table 6. So it can be seen that the

mosquito coil gives the highest risk among all the sources (350 in a million, M₂), followed by the incense sticks (25 in a million, I₁), with the least risk coming from the dhoop. Although this is a preliminary analysis and these numbers are of merely indicative nature, the presence of significant amount of cadmium in one brand of mosquito coil does warrant more quantitative analysis with larger number of brands being tested. Also, the sources for these metals need to be investigated in future studies. One major limitation of this study is lack of quantitative information regarding the different polycyclic aromatic hydrocarbons (PAHs) that might be present in the smoke samples. With hope for better analytical resources in the future, this is something at top priority for future analyses.

Conclusions

The size distributions of smoke particles emitted from some common indoor combustion sources found in India were investigated. The mode diameter for smoke emitted from cigarettes and mosquito coils ranged from 150 to 160 nm. Significantly different results were seen in the case of the incense sticks, which gave modes of 79 to 89 nm. Also, it was concluded that except for incense sticks, all sources showed same measurements (from both long and nano DMA) in the common size range of 25 to 160 nm. The dhoop, however, gave a mode at around 240 nm, much higher than the other sources, but quite close to the removal efficiency minimum of 100–400 nm in the efficiency curve, indicating a greater availability in indoor environments. However, the number concentrations at the mode from all these sources were found to be of the same order of magnitude, i.e., $\sim 10^6$ aerosols cm^{-3} , with the densest aerosol being produced by the dhoop, with a number density of 7.1×10^6 aerosols cm^{-3} . The temporal evolution of the aerosol was also studied, which showed that a steady state corresponding to the number distribution was reached quickly. The modes of sources other than the dhoop and the incense sticks showed up at between 100 and 160 nm, which correspond to a maximum depositional efficiency in the lungs. Also, with the mosquito coils and dhoop having the highest burnout time, they will persist for a longer time and contribute in a sustained manner to pollution. Chemical analysis of the collected samples showed that mosquito coils contain the highest amounts of lead, with the least amount being found in cigarettes. The Cd and Zn levels too followed the same trend, although Zn content in all the sources was higher as compared to other metals.

The mosquito coil M₂ showed cadmium concentrations twice that of the Cal EPA REL level. It also showed the highest carcinogenic risk, 350 people at risk per million of the population. The values of zinc reported for the sources were found to be well below the standard limits, implying the least risk. The calculations for $t_{1/2}$ and t_2 show that the incense particles have a higher tendency to persist in the indoor environment. They also have a higher probability to deposit in the lungs, and thereby to cause damage.

Table 5. Comparison of the calculated toxic potential for various metals with the standards.

Metal	Source	Calc	ACGIH TLV	NIOSH REL	NIOSH IDLH	OSHA PEL	LC ₅₀	Cal EPA REL	LOAEL
Cd	C ₁	2.7 × 10 ⁻³	10	9000		100	5 × 10 ⁵	0.1	20
	C ₂	2.3 × 10 ⁻³							
	I ₁	0.015							
	I ₂	0.0044							
	M ₁	0.0044							
	M ₂	0.21							
	D ₁	5.25 × 10 ⁻⁴							
Pb	C ₁	0.034	50		100	50			
	C ₂	0.0065							
	I ₁	0.28							
	I ₂	0.18							
	M ₁	0.3							
	M ₂	0.67							
	D ₁	0.51							
Zn	C ₁	0.66				1000 (A)		35	
	C ₂	0.47				5000 (B)			
	I ₁	1.04							
	I ₂	3.43							
	M ₁	0.36							
	M ₂	0.75							
	D ₁	1.8							

ACGIH TLV - American Conference of Governmental and Industrial Hygienists' Threshold Limit Value, NIOSH REL - National Institute of Occupational Safety and Health's Recommended Exposure Limit, OSHA PEL - Occupational Safety and Health Administration's Permissible Exposure Limit, LC₅₀ - Lethal concentration, Cal EPA REL - California Environment Protection Agency Reference Exposure Level, LOAEL - Lowest Observed Adverse Effect Level, NIOSH IDLH - National Institute of Occupational Safety and Health's Immediately Dangerous to Life and Health.

Table 6. Carcinogenic risk for Cadmium (in numbers per million of population).

Source	Carcinogenic Risk
Cigarette	C ₁ 5 in 10 ⁶
	C ₂ 4 in 10 ⁶
Incense	I ₁ 25 in 10 ⁶
	I ₂ 8 in 10 ⁶
Mosquito	M ₁ 8 in 10 ⁶
Coils	M ₂ 350 in 10 ⁶
Dhoop	D ₁ ~1 in 10 ⁶ (lesser than that of unit cancer risk)

Appendix: Personal exposure assessment and lifetime cancer risk calculations for toxic metals

The main body of this article dealt with the exposure assessment of inhabitants as a whole, in a typical indoor environment. Here in this study, calculations were done using a slightly different approach, and the exposure of an individual to any of the cigarette brands was calculated, and thereby compared to the calculated values of the standards prescribed by the Occupational Safety and Health Administration (OSHA). The lifetime cancer risk for cases in which the data were available was also evaluated. These calculations were performed using the method of Kammen and Hassenzahl (2001). The abbreviations used and their physical significance are given in Table A.1. The assumptions involved in the calculation are as follows:

1. The number of cigarettes smoked by an individual in a day, given by N_D , is assumed to be 15 (Kammen & Hassenzahl, 2001).
2. The total daily inhalation, TDI, is taken to be 22 m³ day⁻¹ (Kammen & Hassenzahl, 2001).
3. An alternative value of TDI, 12 m³ day⁻¹, as given in Nazaroff and Cohen (2004), was also studied.
4. We further assume that an individual smokes 15 cigarettes over 15 waking hours, thereby giving an average of 1 cigarette per hour for N_H , where N_H is the average number of cigarettes smoked per hour.
5. We also assume that a smoker starts at the age of 15, and smokes for the remainder of a 60-yr lifetime, thereby giving 45 yr in a smoker lifetime (SL) (Kammen & Hassenzahl, 2001).
6. The total volume of air inhaled by an individual during his or her lifetime, TVI, is taken to be 4.7 × 10⁸, as per the calculations of Kammen and Hassenzahl (2001). An alternative value of 2.63 × 10⁸ as specified by Nazaroff and Cohen (2004) was also explored.

The calculations are done as follows:

$$TDE = N_D C_s \quad (A.1)$$

where C_s is metal content per unit source and is obtained from Table 3.

$$ADE = TDE / TDI \quad (A.2)$$

$$HE = C_s N_H \quad (A.3)$$

Table A.1. Abbreviations used in appendix.

Abbreviation	Meaning	Physical Significance	Units
TDE	Total daily exposure	It gives the total amount of a substance inhaled by an individual in a day	$\mu\text{g day}^{-1}$
TDI	Total daily inhalation	Total amount of air inhaled by an individual in a day	$\text{m}^3 \text{day}^{-1}$
ADE	Average daily exposure	The average concentration of heavy metal in the smoke to which an individual is exposed to in a day; it is the ratio of TDE to TDI	$\mu\text{g m}^{-3}$
HE	Hourly exposure	It gives the total amount of a substance inhaled by an individual in an hour.	$\mu\text{g h}^{-1}$
HIR	Hourly inhalation rate	Total amount of air inhaled by an individual in an hour.	$\text{m}^3 \text{h}^{-1}$
AHE	Average hourly exposure	The average concentration of heavy metal in the smoke to which an individual is exposed to in 1 h; it is the ratio of HE to HIR.	$\mu\text{g m}^{-3}$
TLI	Total lifetime intake	It is the total amount of a given species that the individual is exposed to in his or her lifetime.	μg
TVI	Total volume inhaled	Total amount of air inhaled by an individual in his or her lifetime	m^3
LAE	Lifetime average exposure	It gives the average concentration of species in the smoke to which the individual is exposed to in a lifetime.	$\mu\text{g m}^{-3}$
SL	Smoker lifetime	Longevity of a smoker	yr
URF	Unit risk factor		$(\mu\text{g m}^{-3})^{-1}$

Table A.2. Risk parameters evaluated using the TDI value given in Kammen and Hassenzahl (2001).

Metal	Source	TDE	ADE	HE	AHE	TLI	LAE	LCR
Pb	C ₁	1.8	0.082	0.12	0.013	29,565	0.063	
	C ₂	0.36	0.016	0.024	0.027	5913	0.013	
Cd	C ₁	0.134	0.0061	0.0089	0.0097	2193	0.0047	8.4×10^{-4}
	C ₂	0.12	5.5×10^{-4}	0.008	0.0087	1971	0.0042	7.6×10^{-4}
Zn	C ₁	345	15.68	23	25.56	5.7×10^6	12.06	
	C ₂	24	1.1	2	1.78	3.94×10^5	0.84	

Table A.3. Risk parameters evaluated using the TDI value given in Nazaroff and Cohen (2004).

Metal	Source	TDE	ADE	HE	AHE	TLI	LAE	LCR
Pb	C ₁	1.8	0.15	0.12	0.24	29,565	0.11	
	C ₂	0.36	0.03	0.024	0.048	5913	0.023	
Cd	C ₁	0.134	0.011	0.0089	0.018	2193	8.34×10^{-3}	1.5×10^{-3}
	C ₂	0.12	0.01	0.008	0.016	1971	7.5×10^{-3}	1.35×10^{-3}
Zn	C ₁	345	28.75	23	46	5.7×10^6	21.56	
	C ₂	24	2	2	3.2	3.94×10^5	1.5	

The hourly inhalation rate (HIR) is calculated by dividing the total daily inhalation by 24. So,

$$\text{HIR} = \text{TDI}/24 \tag{A.4}$$

Therefore,

$$\text{AHE} = \text{HE}/\text{HIR} \tag{A.5}$$

The total lifetime intake is calculated (Kammen & Hassenzahl, 2001) as

$$\text{TLI} = N_D \times 365 \times \text{SL} \times C_s \tag{A.6}$$

Therefore,

$$\text{LAE} = \text{TLI}/\text{TVI} \tag{A.7}$$

So, lifetime cancer risk (LCR) is then calculated as

$$\text{LCR} = \text{URF} \times \text{LAE} \tag{A.8}$$

The unit risk factor for cadmium is $1.8 \times 10^{-3} (\mu\text{g m}^{-3})^{-1}$ (U.S. EPA, 1999).

The results, for the three metals and two different sources, using the TDI value of Kammen and Hassenzahl, 2001, are given in Table A.2.

Similarly, the results for all the three metals and two different sources, using the TDI value of Nazaroff and Cohen (2004), are given in Table A.3.

From Table A.2 and A.3 we can see that the TDI of a person smoking brand C₁ is higher than for one smoking C₂. This is true for all metals under consideration, and is obvious, since the heavy metal content in C₁ is greater than that in C₂. Therefore, similar results for C₁ vis-à-vis C₂ are also seen for the other parameters evaluated. The values of ADE, AHE, and LAE as calculated by using the TDI value of Nazaroff and Cohen (2004) are roughly twice those obtained using Kammen and Hassenzahl (2001), which could be explained by the fact that the flow rate of former is roughly half that of the latter. The brand C₁ also gives a slightly higher LCR than C₂, implying that the probability of an individual smoking C₁ is slightly greater than C₂.

We can further see from Table A.2 and Table A.3 that none of the lead concentrations so calculated, either on an hourly or on a daily basis, using either value of flow rates, exceeds the OSHA standard of $50 \mu\text{g m}^{-3}$. The same could be said regarding cadmium (OSHA standard $100 \mu\text{g m}^{-3}$) and zinc (OSHA standard $5000 \mu\text{g m}^{-3}$).

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