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Fine particle number and mass concentration measurements in urban Indian households

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Abstract

Fine particle number concentration (D_p>10 nm, cm^{-3}), mass concentrations (approximation of $PM_{2.5}$, μg m^{-3}) and indoor/outdoor number concentration ratio (I/O) measurements have been conducted for the first time in 11 urban households in India, 2002. The results indicate remarkable high indoor number and mass concentrations and I/O number concentration ratios caused by cooking. Besides cooking stoves that used liquefied petroleum gas (LPG) or kerosene as the main fuel, high indoor concentrations can be explained by poor ventilation systems. Particle number concentrations of more than 300,000 cm^{-3} and mass concentrations of more than 1000 μg m^{-3} were detected in some cases. When the number and mass concentrations during cooking times were statistically compared, a correlation coefficient r>0.50 was observed in 63% of the households. Some households used other fuels like wood and dung cakes along with the main fuel, but also other living activities influenced the concentrations. In some areas, outdoor combustion processes had a negative impact on indoor air quality. The maximum concentrations observed in most cases were due to indoor combustion sources. Reduction of exposure risk and health effects caused by poor indoor air in urban Indian households is possible by improving indoor ventilation and reducing penetration of outdoor particles.

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1. Introduction

Understanding human exposure to indoor aerosols and health effects caused by poor indoor air is

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important to enable exposure control and reduction. In the last decade, a number of quantitative epidemiological studies have been carried out in developing countries that attribute specific diseases to the use of solid fuels. This is the case especially for adult women and young children, who receive the highest exposures because of their household roles. In India alone, it has been estimated that the use of biomass fuels causes 400,000–550,000 premature deaths annually among women and children under 5 (Smith, 2000).

There are several ways to reduce pollutant emissions and thereby reduce pollution exposure among populations who cook daily with biomass fuels in developing countries. One way is to improve cooking methods and use cleaner fuels. Even though the use of improved biomass cooking stoves and cleaner fuels (e.g. liquefied petroleum gas (LPG) and kerosene) can improve indoor ambient air quality compared to traditional open fire techniques (Albalak et al., 2001), very little is known about their impact on urban households. Besides cooking techniques, proper ventilation plays an important role. Most cooking appliances are typically not the major contributors to indoor air quality problems. However, the use of unvented appliances, those that are malfunctioning or improperly installed, can generate severe problems (Nagda et al., 1996).

Even though there has been increased interest recently in indoor number concentration as well as the indoor/outdoor (I/O) number concentration ratio (Morawska et al., 2001, 2003; Koponen et al., 2001; Long et al., 2000; Wigzell et al., 2000), the indoor air quality data available today on developing countries are solely based on mass concentration measurements. Many of the recent studies on I/O ratio of the particle number concentration have focused on relatively clean environments, in which the ratio indicates transport of pollutants from outdoor to indoors. In these cases, the value of the ratio is between 0 and 1. However, in indoor environments with strong aerosol sources, the I/O ratio regularly exceeds unity and therefore indicates indoor air quality is dominated by contribution of the indoor sources rather than transport from outdoors. Advanced cooking techniques reduce indoor mass concentration, but these techniques and cleaner fuels may still produce significant amounts of fine particles (D_p>10 nm) and gases, e.g. CO and NO_x (Dennekamp et al., 2001). The control and reduction of fine particle number concentration in ambient air quality is becoming more important, because recent studies have indicated that number concentration could be a better predictor and indicator of the health risks than mass concentration (Donaldson et al., 2001; Peters et al., 1997; Oberdörster, 1995).

In order to extend our understanding of the impact of fine aerosol pollution on human health in India, further information is needed on exposures in terms of metrics such as number concentration, and how these relate to more conventional exposure metrics. In this paper we present data from two studies (one house from the city of Nagpur and 10 houses from the city of Mysore) that involved real-time measurements of aerosol number concentration in the indoor environment. The two datasets also provide information on correlations between number concentration and CO, and number concentration and mass concentration. The objectives of presenting these data together are to: (1) define seasonal and diurnal variations of fine particle (D_p>10 nm) indoor and outdoor number concentration; (2) quantify the impact of indoor activities on indoor particle concentration levels based on real-time measurements and time–activity information; (3) experimentally investigate the relationship between indoor and outdoor particle number concentration in households using LPG or kerosene as the main cooking fuel; (4) determine the real-time diurnal indoor fine particle number and mass (approximation of $PM_{2.5}$) concentration in urban Indian households; and (5) to compare indoor and outdoor number concentration to the CO concentration (ppm).

2. Experimental methods

2.1. Sampling sites and houses

The sampling sites were located in the cities of Nagpur and Mysore. Nagpur is the second capital of the western Indian state of Maharashtra. The city is situated on the banks of Nag River from which it derives its name. It is almost at the geographical centre of India (approximately 79°E, 21°N). It lies on the intersection of the Mumbai–Calcutta and the Delhi–Chennai air, road and rail routes. The surround-

ing region is an undulating plateau rising northward to the Satpura Range, from 271 to 653 m high. The city has a population of about (1991 prelim.) 1.6 million. The measuring site in Nagpur was located in a private flat in a residential area about 3 km east from the city's center called Sitabuldi. The flat consisted of three rooms and a kitchen with an area about 90 m^2. The flat had three air conditioning systems (window units), one placed in each room. These units recycled and cooled the air inside the flat. Otherwise there was no other ventilation system. Exchange of the indoor and outdoor air was only possible through the several gaps (doors and window frames). The number of residents was three and there were no other non-cooking combustion sources in the flat. A layout of the flat is presented in Fig. 1.

The other city of our study, Mysore, is a South Indian city (approximately 76°E, 12°N) situated at a height of 770 m from sea level in the state of Karnataka. It is a small city with a population of 740,000 (2001 Census) and an area of more than 40 km^2. It is about 140 km to the southwest of Bangalore, the capital of the state. The city contains many small businesses, with some small industries, and it is surrounded by small villages, moderate forests, and hill ranges. Vehicle emissions are the main sources of outdoor air pollution in urban areas, while cooking activities with poor ventilation facilities in many of the houses are the main sources of indoor $PM_{2.5}$ pollution. Mass and number concentration measurements in the city were performed indoors in 10 houses from around the city. Houses were chosen from those using kerosene or LPG as the main fuel. Five individual houses of each type were selected from around the city. Kerosene and LPG houses were chosen in pairs such that each kerosene house was in an area close to a LPG house.

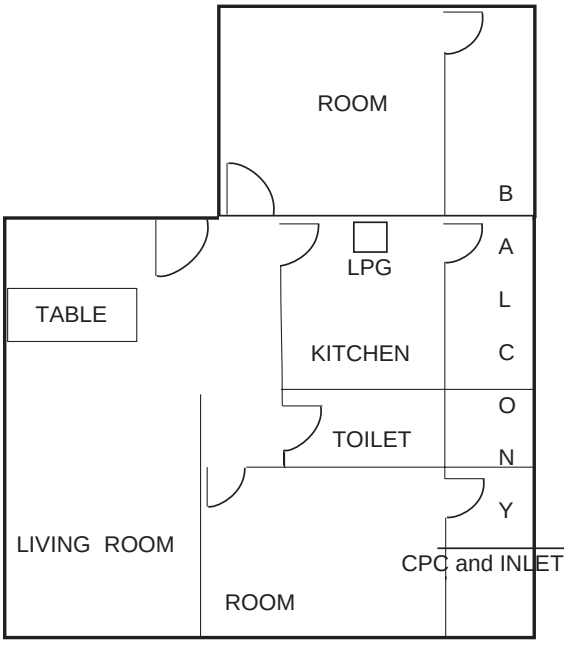

Fig. 1. A layout of the residential house (flat) in Nagpur. During the measuring period in April the CPC located in the table in the living room. The toaster located next to the table above the fridge.

2.2. Instrumentation

The number concentration of the particulate matter was measured with the portable TSI Condensation Particle Counter (CPC 3007, particle size >10 nm). The CPC measurements were continuous, with a time resolution of 1 min. From these data 1 h mean concentrations were calculated for the analysis. The CPC used in Nagpur was calibrated before the measuring period (Hämeri et al., 2002). While measuring the outside concentration, the inlet for the CPC was placed outside, 50 cm from the wall. During indoor measurements the CPC was placed in the room next to the kitchen 2 m above the floor level. The room was located 4 m away from the kitchen (see Fig. 1). During the measuring period in April, the CPC was located on the table in the living room 3 m away from the kitchen door.

CO concentrations were measured using a portable OMNI 4000 gas detector (Enmet). Readings with a time resolution of 1 min were taken for the comparison of indoor and outdoor number concentrations at the end of April and the beginning of May. Temperature and relative humidity were measured using a wet and dry bulb thermometer every hour. Indoor, outdoor temperature and relative humidity data during each measuring period in Nagpur are presented in Fig. 2.

Mass and number concentration were measured simultaneously with a Dust Trak (Model 8520, TSI, particle size 0.1–2.5 μm) and a portable Condensation Particle Counter (CPC 3007, particle size>10 nm) in parallel. Gravimetric $PM_{2.5}$ measurements were also carried out at the same time and location

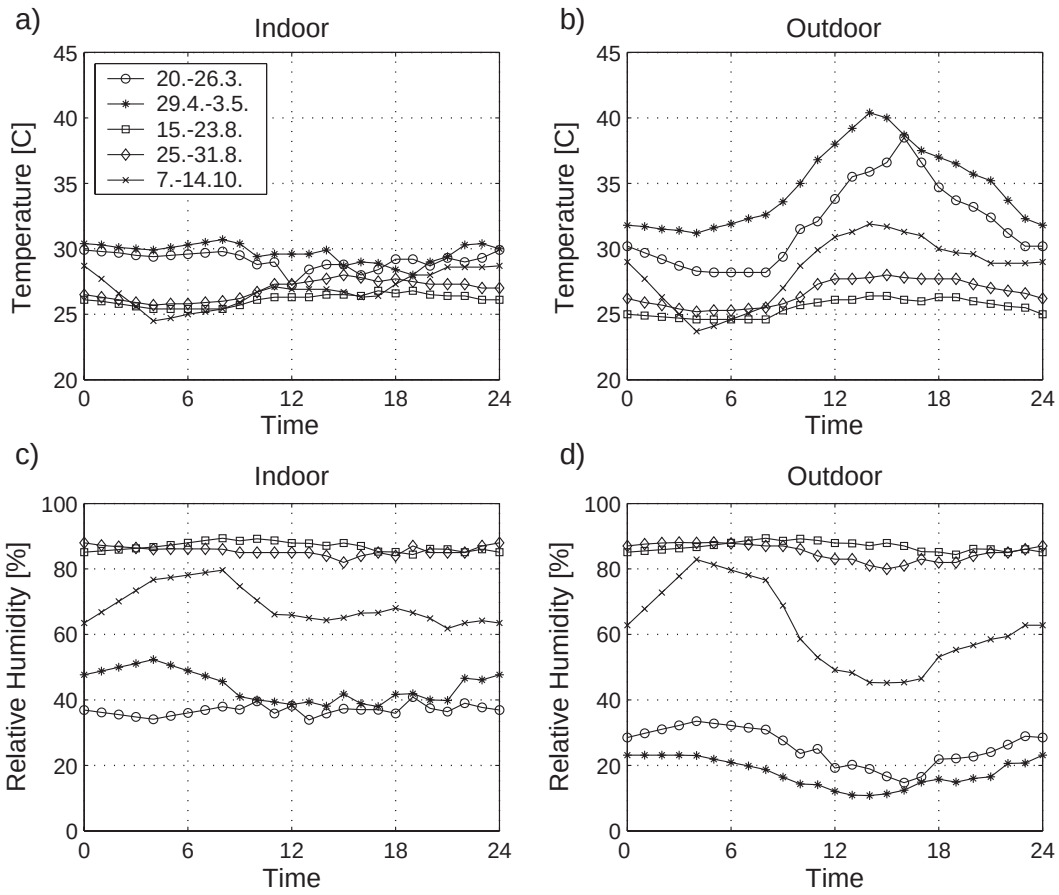

Fig. 2. Diurnal indoor (a and c) and outdoor (b and d) temperature (°C) and relative humidity (%) of the sampling site in different measuring periods in Nagpur 2002.

using a $PM_{2.5}$ Personal Environmental Monitor (PEM™, MSP, USA) operated at 10 l/min. Whatman PTFE membrane filters were used as collection substrates. Filters were pre- and post-weighed after conditioning in the weighing area, and measured aerosol concentrations were adjusted for weight changes in unexposed filters used as field blanks. Gravimetric $PM_{2.5}$ measurements made concurrently with co-located real-time DustTracks were used to calibrate measurements made using the real-time mass concentration monitors. For the indoor study, both measurements were continuous with a time resolution of 5 s.

For indoor measurements, the instruments were kept in the living room usually on a table at a height of about 50 cm from the floor. As particles smaller than 2.5 μm aerodynamic diameter were being measured, it was assumed that even slight air movements within the room would lead to turbulent mixing, resulting in a well-mixed aerosol. Air was aspirated through short lengths of plastic tubing connected to the inlets of the instruments. For outdoor measurements, the plastic tubing protruded 20 cm from the building wall into the ambient air. Electrostatic and diffusional losses are always a concern when sampling sub-micrometer particles. In this instance, although the plastic tubing may have held an electrostatic charge, it was assumed that the sampled aerosol would be at charge equilibrium, thus leading to relatively small electrostatic losses. The length and diameter of the tubing, together with the sampling flow rate, will have led to estimated

losses lower than 5% for 10 nm diameter particles, and 0.5% for 100 nm diameter particles (Gormley and Kennedy, 1949) Temperature and relative humidity were noted during the particle and CO measurements.

The experiments were performed in March, April, May, August and October in the city of Nagpur and in August, October and December at several locations in the city of Mysore in 2002. The duration of the experiments varied from a few days to 2 weeks in each measuring period. In Nagpur, the CPC was used 24 h during each measuring period. The CPC 3007 required propanol to be added every 4–5 h. Propanol was added before the efficiency of CPC decreased to ensure high quality data.

A summary of the measuring periods and equipment is shown in the Table 1. The I/O number concentration ratio was defined by measuring 1 h indoors and then the subsequent hour outdoors. The measured indoor and outdoor hours varied every day, i.e., if on Tuesday indoor number concentration was measured between 1 and 2 p.m., on Wednesday the outdoor number concentration was measured at the same time. With this approach, we were able to minimize the possible effect of diurnal patterns. At the end of April and the beginning of May, indoor and outdoor particle number concentrations were measured using a different approach. First we measured 7 days only indoors and then 5 days outdoors. The time resolution during these measurements was also 1 min. Hence, I/O number concentration ratio could not be defined during this time period.

Measurements in Mysore were carried out in five households that used LPG and five households that used kerosene as a main cooking fuel. All measuring periods were conducted between August and December 2002. For sampling, all the above mentioned instruments were taken to the sampling location and run from a 12 V battery for 24 h. The measuring period was repeated in the same households after

Table 1
The summary of the measuring periods, main fuel type, parameters measured, time resolution and equipments used in this study in the city of Nagpur and Mysore, India 2002

Place	Measuring period	Main fuel type	Parameters measured	Resolution	Equipments
Nagpur	20.–28.3.2002	LPG	Number concentration (cm^{-3})	1 min	TSI CPC 3007
	22.–29.4. (indoor only)	LPG	CO concentration (ppm)	1 h/1 min	OMNI 4000
	29.4.–3.5. (outdoor only)	LPG	Temperature (°C)	1 h	Wet/dry bulb thermometer
	15.–23.8.	LPG	Relative Humidity (%)	1 h	
	25.–31.8.	LPG			
	7.–14.10.	LPG			
Mysore	August	LPG	Mass concentration ($\mu g\ m^{-3}$)	5 s	TSI DustTrak 8520
	October	LPG	Number concentration (cm^{-3})		
	October	LPG			
	October	LPG		5 s	TSI CPC 3007
	October	LPG			
	December	LPG			
	December	LPG			
	December	LPG			
	December	LPG			
	December	LPG			
	August	Kerosene			
	October	Kerosene			
	October	Kerosene			
	October	Kerosene			
	December	Kerosene			
	December	Kerosene			
	December	Kerosene			
	December	Kerosene			
	December	Kerosene			

about two months. The responsible family member in the house was asked to note down the timings and details of combustion activities over the day. Of the households visited, four used coconut husks for heating water (two LPG and two kerosene). This activity occurred every day early morning from around 5 to 8 a.m., and went on for around 2 to 3 h. Aerosol generation from burning incense sticks (about 2 at a time) was common. The duration was, about 15 min, two times a day—morning and evening. Wherever possible, the CPC was recharged with alcohol to obtain longer sampling times, although it was frequently not possible to obtain complete 24 h exposure records. Both mass and number concentrations were recorded as 5-s averages. These data were synchronised, and averaged over a 1-min rolling period.

3. Results and discussion

3.1. Indoor and outdoor number concentrations

Fig. 3a presents 1 h mean indoor number concentrations measured during different seasons in Nagpur. The indoor number concentrations were at minimum during the night (from 11 p.m. to 6 a.m.). The peak value early in the morning in April was

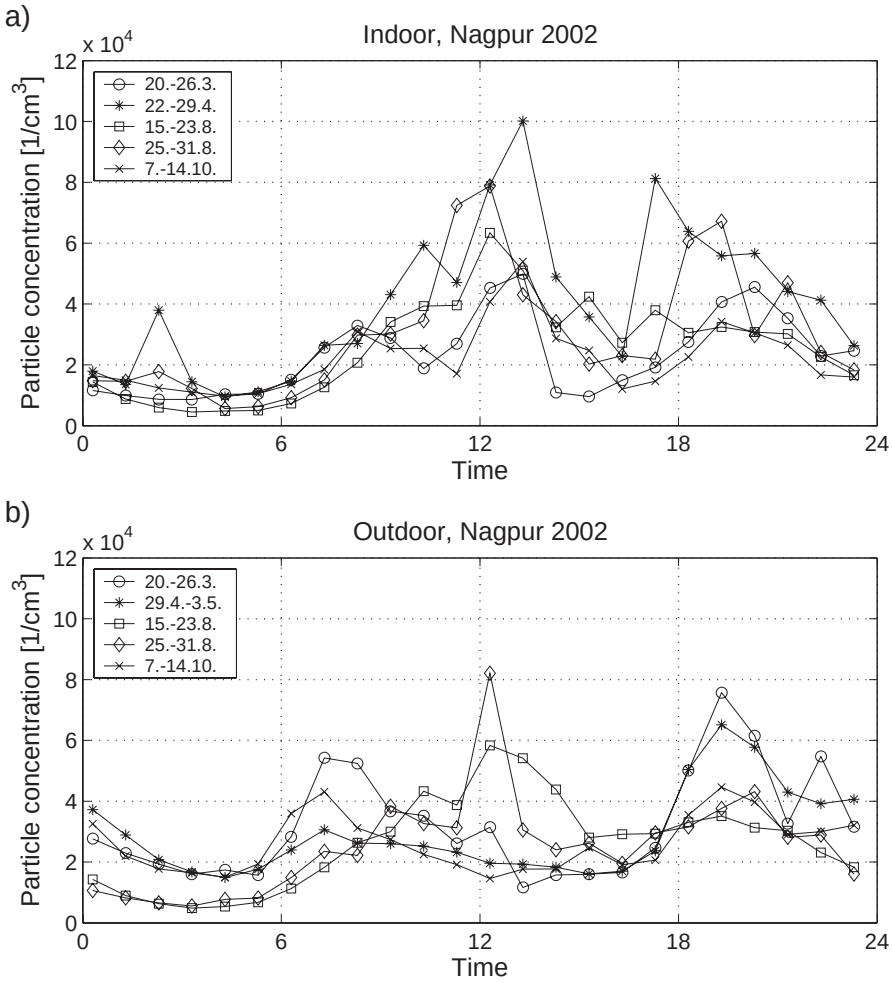

Fig. 3. Diurnal 1 h mean indoor (a) and outdoor (b) particle concentrations (cm^{-3}) in different measuring periods in Nagpur 2002.

result of nighttime cooking (a toaster) during the measuring period (see, e.g. Fig. 4a, day #113 and day #115). The maximum number concentration values were observed around 12 a.m. and 6 p.m. The maximum 1 h mean was 101,000 particles cm^{-3} (in April, at 1.30 p.m.) and the minimum was 4400 particles cm^{-3} (in August, 3.30 a.m.). In Nagpur only LPG was used for cooking.

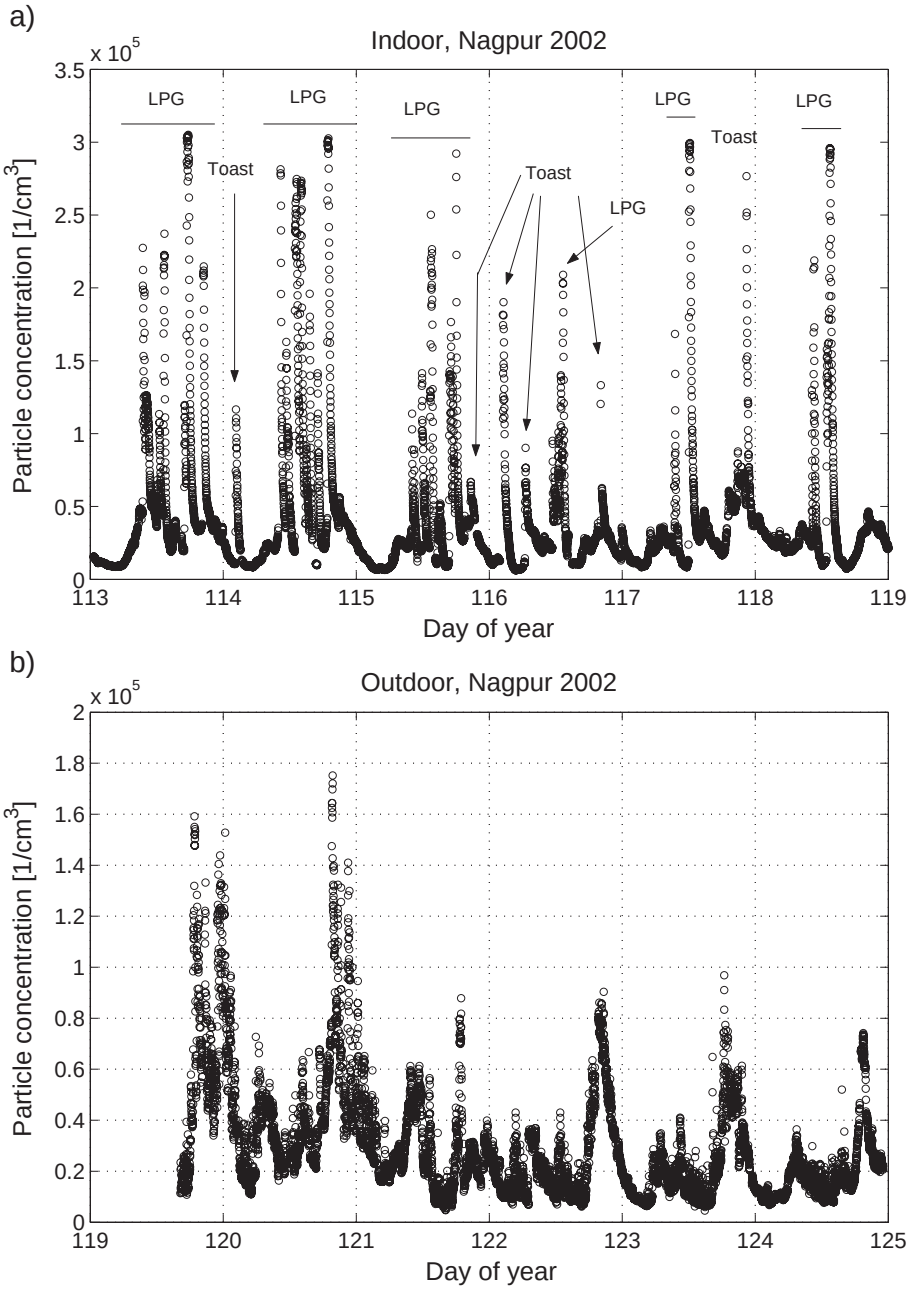

Fig. 4. Diurnal indoor (a) and outdoor (b) particle concentrations (cm^{-3}) with the time resolution on 1 min in April in Nagpur 2002. LPG=cooking with liquefied petroleum gas; Toast=use of toaster.

High 1 h mean indoor number concentrations can be explained by a strong source generating the particles or by poor ventilation or a combination of the two. Typical Indian urban houses have large fans placed on the ceiling to circulate indoor air. In a kitchen a small fan might be placed in the window to aid circulation. Hence, ventilation only occurs through open windows or doors. During hot seasons, wealthy people might use air conditioning (AC), typically window units.

Fig. 3b presents the 1 h mean outdoor number concentrations measured during different seasons in Nagpur. Concentrations were lowest during night (12 p.m.–5 a.m.). After 6 a.m. the concentrations increased. This increase was most probably due to the morning vehicle traffic peak hours (7–9 a.m.). Also, the increase after 6 p.m. can be assumed as a result of vehicle traffic, but the impact of biomass and/or refuse burning cannot be neglected (Sharma et al., 2003; Mänkkönen et al., 2004b). Similar fluctuations in number concentration of ambient air were also observed in New Delhi (Mönkkönen et al., 2004a).

In August (15.–23.8. and 25.–31.8.) number concentrations were lowest and no clear morning peak hours were observed. During these two measuring periods the relative humidity was very high due to heavy monsoon rains. However, high number concentrations right after noon were observed. There is

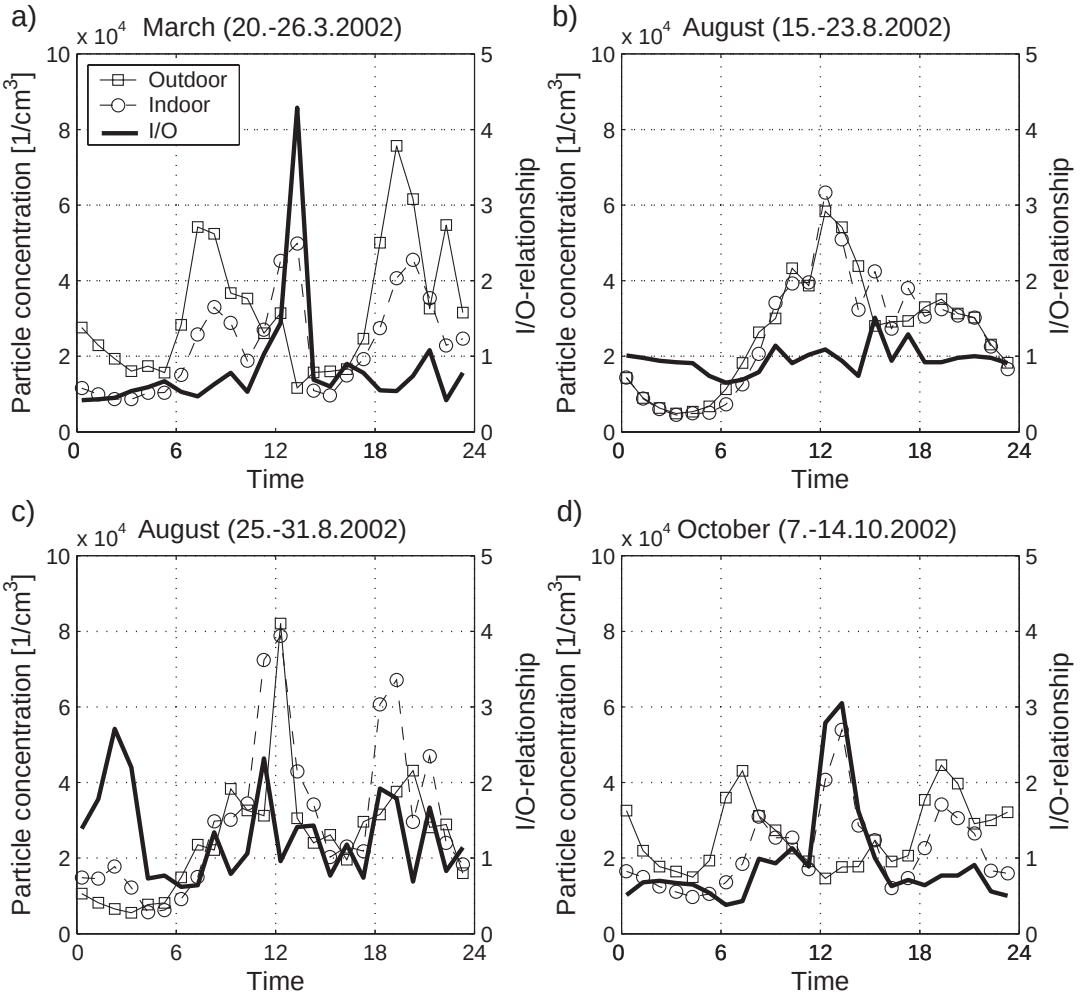

Fig. 5. Diurnal 1 h mean indoor and outdoor number concentrations (cm^{-3}) and I/O relationship in different seasons in Nagpur 2002.

no clear explanation for this peak. The most evident reason is that the particles formed during the cooking process were transported outside through the open kitchen door. The door from the kitchen to the balcony was constantly open during this measuring period to cool the kitchen. The door was usually closed, since the flat had air conditioning (a window unit) system. In August the air conditioning was not used because the outside temperature was sufficiently low. This speculation is also supported by comparing Fig. 3a and b. First the indoor number concentration increased and after about 1 h also the outdoor number concentration increased with the same magnitude. The inlet of the CPC located at the same side as the balcony 3 m away from the balcony.

The indoor mean particle number concentration (24 h average) in March was 23,000 cm^{-3}, in April 41,000 cm^{-3}, in August 25,600 cm^{-3} and 30,400 cm^{-3} and in October 22,000 cm^{-3}. The outdoor 24 h mean number concentrations were 32,300 cm^{-3}, 29,200 cm^{-3}, 26,200 cm^{-3}, 25,300 cm^{-3} and 26,100 cm^{-3}, respectively. The 24 h mean indoor number concentration was slightly higher only in the second measuring period in August. The indoor/outdoor dataset measured in April is not as directly comparable as for the other seasons since the indoor and outdoor concentrations were measured during different weeks.

Real-time particle concentration data combined with the human activities allow the sources of particle diurnal variation to be analyzed. The impact of human activities to the number concentration can be seen well in Fig. 4a. This figure presents the indoor number concentration with the time resolution of 1 min, measured in April. High concentrations up to 300,000 particles cm^{-3} were observed. The peaks around noon, afternoon and evenings were clearly caused by the cooking with LPG. Few clear peaks (100,000 $cm^{-3}<N<280{,}000$ cm^{-3}) caused by the use of the toaster were observed early in the morning (day #114 and day #116) and at 9 a.m. (day #116) and in the evening (day #115, 10 p.m., day #116, 10 p.m. and day #117, 11 p.m.). The toaster was located above the refrigerator 1.5 m above the floor level and 1.5 m away from the table (CPC). A similar connection of human activities to the number concentration has also been found by Morawska et al. (2003) and Patterson and Eatough (2000).

3.2. Indoor–outdoor relationship

Fig. 5 presents the indoor/outdoor (I/O) number concentration relationship of the number concentration in each measuring period. The minimum 1 h mean I/O number concentration ratio was 0.38 (observed in October, 6.30 a.m.) and the maximum was 4.29 (observed in March, 12.30 a.m.). The highest 24 h geometric mean I/O number concentration ratio 1.17 was recorded in August (25.–31.8., Fig. 5c) and the lowest 0.70 in March. The 24 h geometric mean I/O number concentration ratio during the first measuring period in August (15.–23.8., Fig. 5b) was 0.94. As considered earlier, the reason for this high I/O number concentration ratio during both measuring periods in August is that doors from the flat to the balcony were open most of the time, since there was no need to use air conditioning system due the low outdoor temperatures. As the result of open doors, the indoor and outdoor concentrations followed quite well with each others. The 24 h geometric mean I/O number concentration ratio in October was 0.80. Table 2 summarizes the minimum and maximum indoor and outdoor number concentrations measured in Nagpur.

Only few studies have reported the I/O number concentration ratio in the literature. Koponen et al. (2001) found that indoor particle concentrations were about the times smaller that the outdoor concentrations in an office building near Helsinki downtown. Tu and Knutson (1988) used an electrical aerosol analyzer (EAA) to measure I/O ratio in single family houses. In these houses the I/O ratio varied from 0.2 to 41 depending strongly on indoor human activities.

Table 2
The summary of the 1 h mean, minimum and maximum indoor and outdoor particle number concentration ($cm^{-3}\times1000$) measured in different seasons in Nagpur 2002

One hour mean indoor and outdoor number concentrations during different seasons in Nagpur										
	March		April		August		August		October	
	In	Out	In	Out	In	Out	In	Out	In	Out
Average	23.1	32.3	40.7	30.0	25.6	26.2	30.4	28.3	22.0	26.2
S.D.	13.0	17.2	24.3	13.8	15.9	15.1	21.0	16.4	10.8	9.1
Max	49.9	61.6	100.2	65.1	63.4	58.4	78.9	82.1	53.9	44.6
Min	8.6	11.6	9.5	14.9	4.5	4.9	5.6	5.6	9.7	14.6

3.3. Indoor number vs. mass concentration

Fig. 6 presents an example of the indoor number concentration and mass concentration measured in sites where LPG (Fig. 6a) or kerosene (Fig. 6b) was used as a cooking fuel. Clear peaks both in number and mass concentration caused by cooking were observed during the afternoon in both measuring

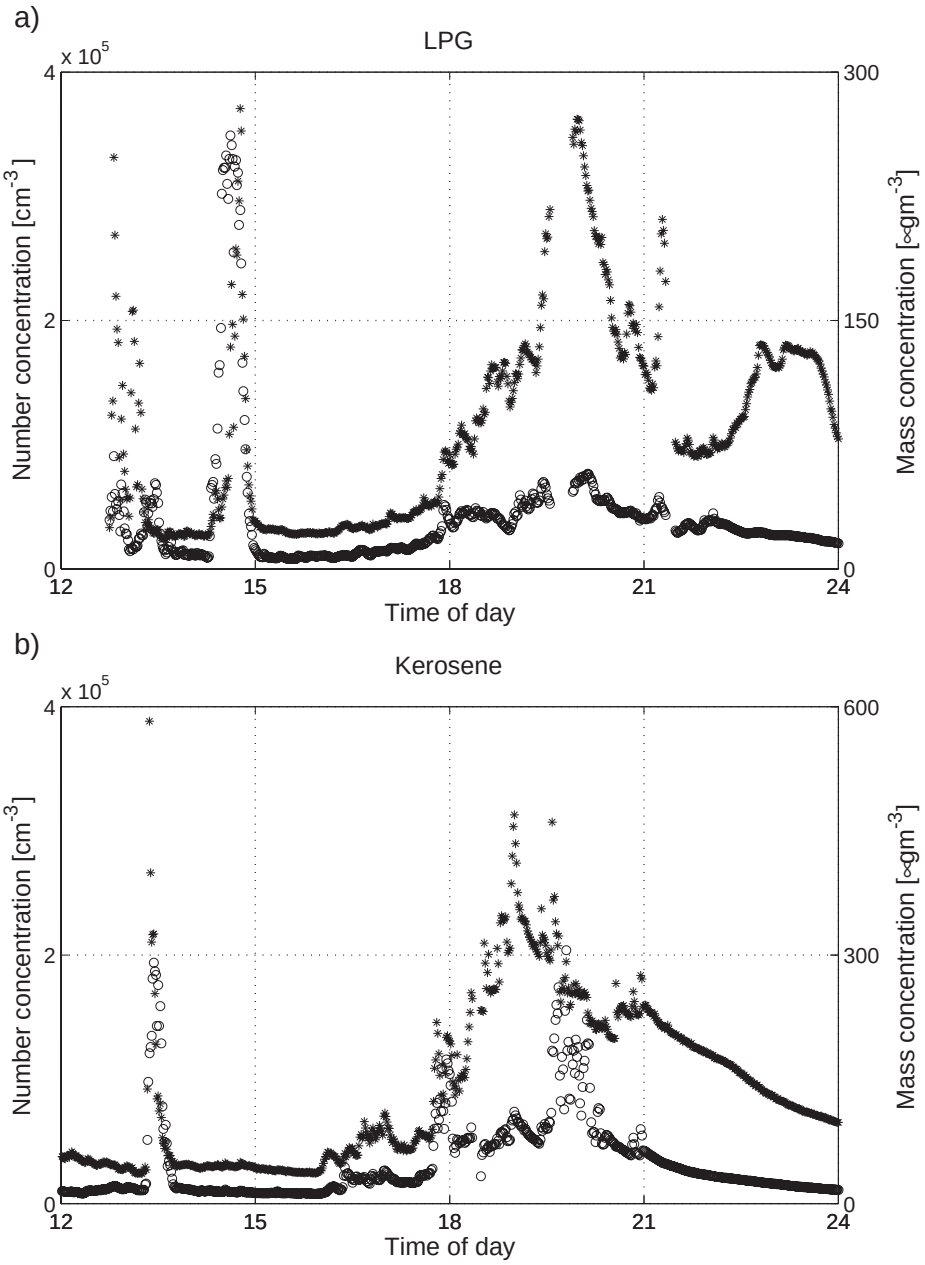

Fig. 6. An example of a diurnal indoor number (○, cm^{-3}) and mass (*, $\mu g\ m^{-3}$) concentration with the time resolution of 1 min in the measuring sites that used LPG (a) and kerosene (b) as a cooking fuel.

sites. From the figures we can also see that the number and mass concentration values followed well with each other in the afternoon but toward the evening (after 6 p.m.) a greater increase in mass concentration compared to number concentration was observed. This is most likely explained by other sources and normal living activities, or by a change in the number size distribution. In India, families return back to home usually after 5 p.m. which might generate indoor particles. The penetration of outdoor dust particles also cannot be neglected.

The mean number and mass concentrations, standard deviations of the concentrations and correlation coefficient (between mass and number concentrations) during each measuring period and site in Mysore is presented in the Table 3. Table 3a and c presents the calculated results for all the measured data while Table 3b and d presents only the measured data during cooking processes. As the CPC did not run continuously for 24 h in many cases, correlations between overall mass and number concentration measurements will be affected by differences in measurement periods, particularly where major events occurred when the CPC was not running. Isolating the comparison to cooking events where both instruments were operating provides a clearer indication if the correlation between number and mass concentrations. The correlation coefficient for all the measured data varied from 0.87 to −0.29 and from 0.92 to −0.30 during the cooking processes. High variations in correlation coefficient were also found by Morawska et al. (2003). In their study, average correlation in

Table 3
Indoor mean mass concentration (μg m^{-3}), number concentration ($1000\times cm^{-3}$) and correlation coefficient in different measuring sites using LPG (sites 1–5) or kerosene (sites 6–10) as main cooking fuel

House ID	No. of Residents	m (μg m^{-3})		N ($1000\times cm^{-3}$)		r	
		1st set	2nd set	1st set	2nd set	1st set	2nd set
(a) Indoor mass and number concentrations and correlation coefficient for all the results (LPG as main cooking fuel)							
1	3	51.0 (33.3)	27.0 (21.4)	21.4 (10.7)	22.1 (9.1)	0.869	0.326
2	3	64.2 (171.3)	56.7 (52.1)	14.2 (12.7)	13.6 (11.4)	0.651	0.421
3	3	116.4 (129.3)	48.2 (87.1)	34.5 (47.6)	14.0 (10.5)	0.320	−0.218
4	4	94.6 (124.4)	146.1 (292.6)	32.5 (29.1)	79.3 (79.6)	0.431	0.789
5	4	162.6 (362.4)	69.5 (101.5)	11.9 (10.5)	20.5 (21.8)	0.734	0.263
(b) Indoor mass and number concentrations and correlation coefficient during cooking process (LPG as main cooking fuel)							
1	3	84.3 (43.0)	34.5 (25.0)	23.0 (12.9)	32.1 (9.1)	0.917	0.192
2	3	254.2 (429.5)	74.1 (58.6)	27.0 (23.7)	32.6 (16.3)	0.681	0.187
3	3	102.5 (122.0)	151.7 (170.4)	27.6 (27.7)	18.5 (13.0)	0.305	−0.300
4	4	142.2 (93.9)	261.2 (392.0)	89.6 (12.5)	125.6 (96.0)	0.869	0.766
5	4	565.2 (731.3)	29.7 (1.0)	32.1 (8.9)	35.6 (17.4)	0.832	−0.032
(c) Indoor mass and number concentrations and correlation coefficient for all the results (kerosene as main cooking fuel)							
6	3	96.9 (84.8)	–	27.4 (35.4)	–	0.112	–
7	3	107.9 (87.2)	121.1 (321.0)	26.3 (30.8)	64.1 (69.0)	0.165	0.172
8	3	146.2 (162.5)	41.0 (42.1)	43.1 (36.7)	23.8 (9.4)	−0.058	−0.272
9	5	63.7 (62.7)	52.1 (97.1)	22.6 (20.0)	26.2 (37.2)	−0.288	0.145
10	3	106.7 (153.0)	67.0 (92.0)	17.5 (18.2)	252.0 (34.0)	0.068	0.309
(d) Indoor mass and number concentrations and correlation coefficient during cooking process (kerosene as main cooking fuel)							
6	3	217.8 (76.0)	–	83.1 (36.2)	–	0.917	–
7	3	239.7 (82.3)	576.8 (1075)	87.5 (42.9)	183.8 (74.2)	0.647	0.320
8	3	187.0 (111.0)	93.8 (58.7)	64.7 (21.1)	–	0.762	–
9	5	67.7 (22.4)	136.4 (150.1)	18.8 (3.3)	47.7 (47.4)	0.715	0.516
10	3	230.7 (150.6)	140.6 (193.1)	42.9 (20.5)	92.0 (38.9)	0.883	0.295

(a) and (c) represent all the measured results and (b) and (d) represent results only during the cooking process. Standard deviation is given in the parentheses.

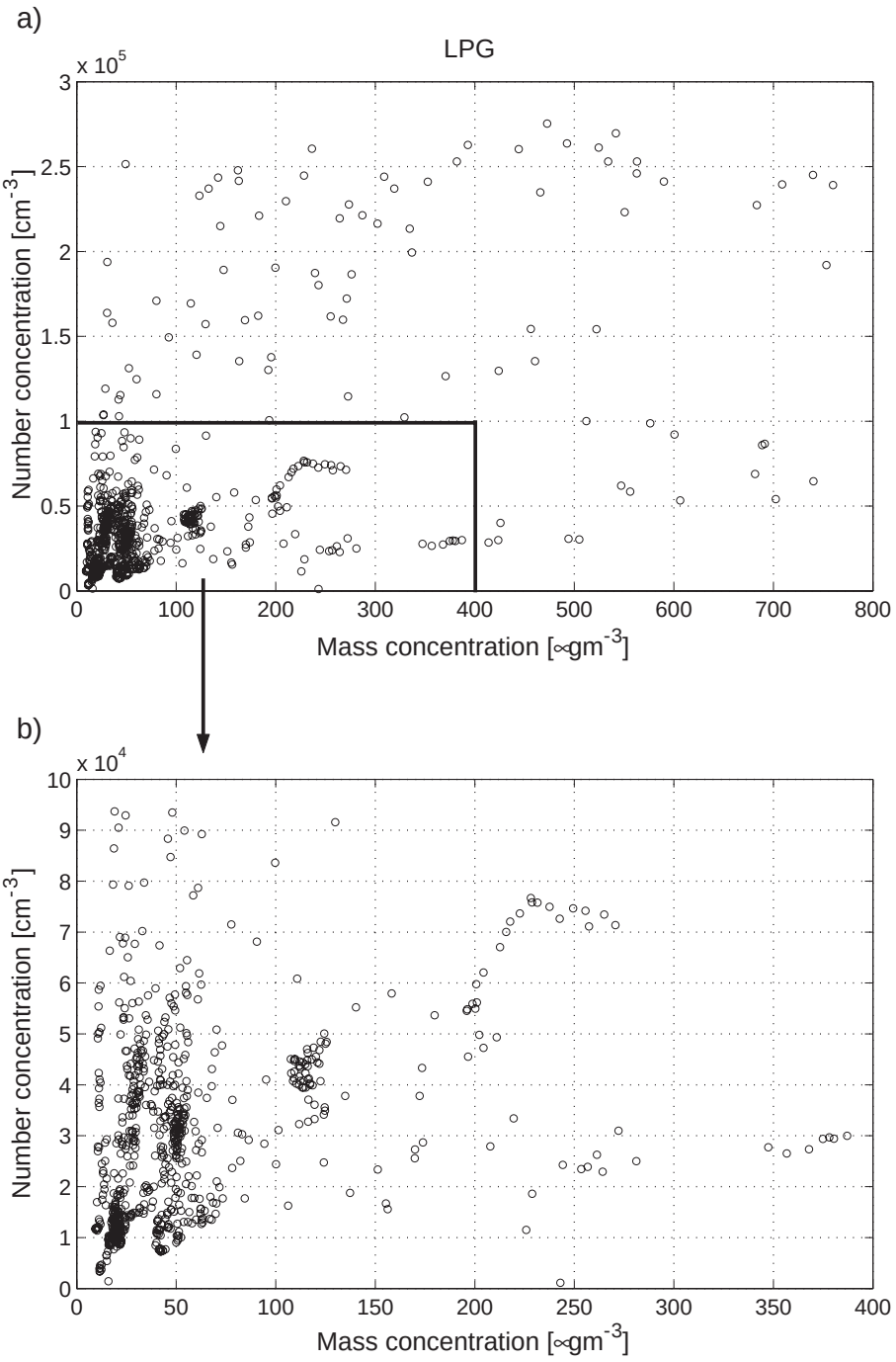

Fig. 7. Indoor fine particle number concentration (D_p>10 nm, cm^{-3}) vs. mass concentration (approximation of PM_1, μg m^{-3}) with the time resolution of 1 min measured during the cooking processes that used LPG as a cooking fuel. (b) is a zoomed fraction of (a).

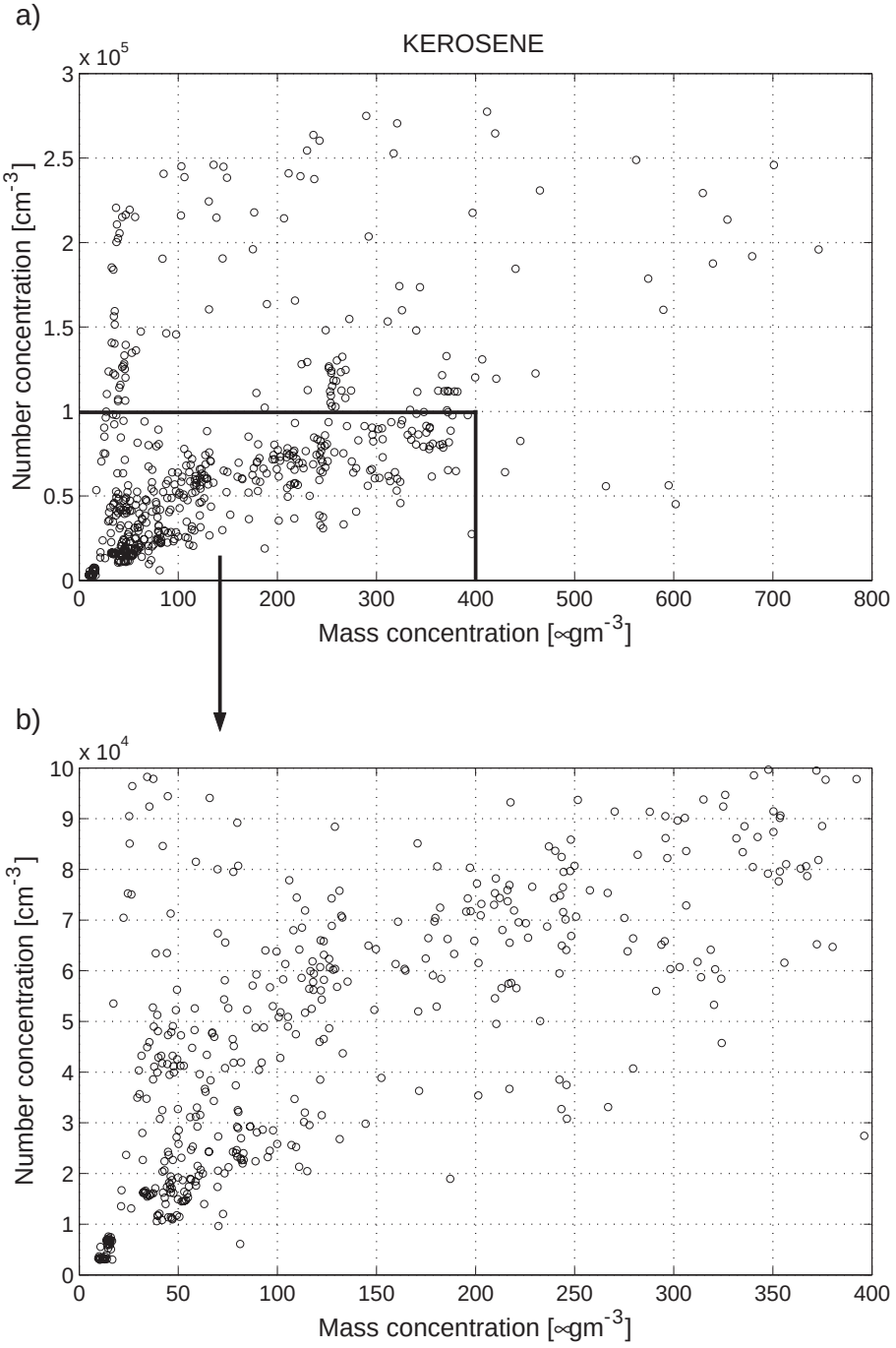

Fig. 8. Indoor fine particle number concentration (D_p>10 nm, cm^{-3}) vs. mass concentration (approximation of PM_1, μg m^{-3}) with the time resolution of 1 min measured during the cooking processes that used kerosene as a cooking fuel. (b) is a zoomed fraction of (a).

residential houses ranged from 0.1 to 0.73 with an overall average 0.48 ± 0.27.

A significant increase in the correlation coefficient compared with all the data vs. cooking data was observed in the measuring sites #4 and #6–10 during the first set and in site #8 during the second set. The fluctuation in the correlation coefficients between different measuring sites can be explained by the different locations of the houses and different living activities in each period. In some cases, the outdoor sources had major effect on the exposure levels indoors. Also the measuring periods were short in each site.

Figs. 7 and 8 present the indoor fine particle number concentration (D_p>10 nm, cm^{-3}) vs. mass concentration (approximation of $PM_{2.5}$, $\mu g\ m^{-3}$) with the time resolution of 1 min measured during all the cooking processes that used LPG (Fig. 7) or kerosene (Fig. 8) as a cooking fuel. The correlation coefficients were 0.57 (LPG) and 0.48 (kerosene), which indicates that other source-related particles were also present in the atmosphere in these households as considered earlier.

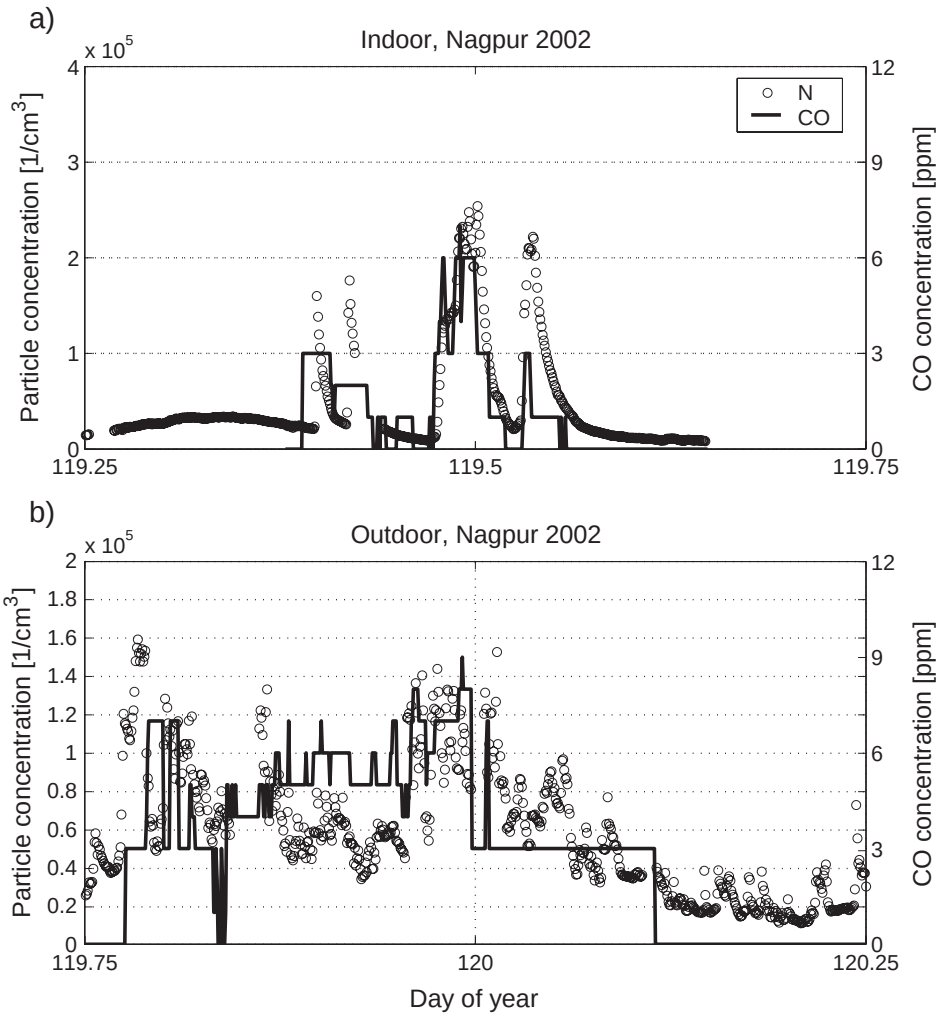

Fig. 9. Diurnal indoor (a) and outdoor (b) particle concentration (cm^{-3}) and CO concentration (ppm) with the time resolution on 1 min in Nagpur, April 2002.

The mean mass and number concentrations increased clearly during the cooking processes. In the sites where LPG was used the mean mass concentration increased from 55.5 μg m^{-3} to 144.6 μg m^{-3} and the mean number concentration increased from 24,000 cm^{-3} to 53,100 cm^{-3}. In the measuring sites where kerosene was used as a cooking fuel, the mass concentration increased from 82.6 μg m^{-3} to 152.3 μg m^{-3} while the mean number concentration increased from 35,500 cm^{-3} to 72,800 cm^{-3}, respectively. The highest measured indoor mass concentration was 3000 μg m^{-3}.

3.4. Number concentration vs. CO concentration

Different combustion processes are widely used for cooking in India. Besides aerosols, combustion processes also produce CO. To evaluate aerosol number concentration exposure levels in Indian houses, cheaper and alternative methods to the CPC are needed. Hence simultaneous number and CO concentration measurements were conducted not only to get a better understanding of the possible aerosol sources but also to find other methods for evaluating aerosol number concentration levels.

Fig. 9a (indoor) and b (outdoor) presents an example of the diurnal number concentration together with CO concentration with time resolution of 1 min measured in the end of April and in the beginning of May. The Fig. 9a time frame is from morning 6 a.m. to evening 6 p.m. and the time frame in Fig. 9b is from evening 6 p.m. to 6 a.m. next morning. As we can see from Fig. 9a, the CO concentration follows the number concentration well. In this figure, all the peaks were caused by cooking with LPG. Even though in this figure the correlation between the number and CO concentration is high, the correlation coefficient for all the data (n=10,056) was only 0.26. There were at least two reasons for this low correlation. Firstly, indoor number concentration was influenced also by other activities than cooking (e.g. cleaning and other living activities), and secondly, the portable gas detector used in the experiment showed only whole number readings. Hence, indoor CO concentration measurements can give a good estimation for number concentration levels if the time resolution is short and the real-time CO concentration data is combined with the human activities. During cooking processes, increases of indoor CO concentration by 1 ppm resulted in a 60,000 cm^{-3} average increase in aerosol number concentration. During the indoor measuring period in April, the 1 h mean CO concentrations were below 5 ppm. Similar 1 h indoor mean CO concentrations have earlier been found by Samet et al. (1987) where gas stoves were in operation.

The correlation coefficient for outdoor number and CO concentration for all the data (n=4903) was 0.73. Especially in the evenings, CO concentration followed well the number concentration. This high correlation indicates that the outdoor particle concentration during the measuring period was strongly contributed by different combustion processes, i.e., burning fossil fuels (traffic and domestic use of LPG and kerosene) and biomass/refuse. This speculation is supported by Sharma et al. (2003). They found that biomass and/or refuse burning are significant contributors to the organic fraction of PM_{10} in New Delhi's atmosphere.

4. Summary and conclusions

Fine particle number (D_p>10 nm, cm^{-3}) and mass concentrations (approximation of $PM_{2.5}$, μg m^{-3}) were measured in a number of urban households in India in 2002. In the city of Nagpur, the number concentration measurements were focused on indoor and outdoor diurnal and seasonal variations and on particle number I/O relationship. In the city of Mysore, the measurements were focused on number concentrations and mass concentrations in the houses that used either LPG or kerosene as a main cooking fuel.

Normal indoor living activities, especially cooking, clearly increased indoor particle number and mass concentration in all measuring sites, but also in some cases the outdoor sources had a major effect on the exposure levels indoors. Indoor number concentrations more than 300,000 cm^{-3} and indoor mass concentrations more than 3000 μg m^{-3} were detected in some cases. Correlation coefficients r>0.50 were observed only in 63% of the cases.

Cooking processes also clearly increased the particle number I/O number concentration ratio and CO concentration. Usually the outdoor concentra-

tions were higher, but I/O number concentration ratios more than 4.00 were detected. During cooking processes, indoor CO concentration closely tracked the number concentration. In the evening, outdoor CO concentration followed the outdoor number concentration. A high correlation (r=0.73) indicates that the outdoor particle concentration during the measuring period was strongly influenced by different outdoor combustion processes, i.e., burning of fossil fuels (traffic and domestic use of LPG and kerosene) and biomass/refuse.

Clear seasonal variations were observed. The indoor number concentration (24 h average) was lowest in October (22,000 cm^{-3}) and highest in the end of April (41,000 cm^{-3}) and the outdoor number concentration was lowest in August (25,300 cm^{-3}) and highest in March (32,300 cm^{-3}), respectively. In March and April the relatively humidity was at minimum and no rains were observed which allowed small dust particles to be resuspended from the soil to atmosphere, hence decreasing the particle number concentration. Low number concentration levels during high relatively humidity periods can be explained by scavenging of the particles.

Despite the cleaner fuels, the cooking stoves that used LPG and kerosene produced significant fine particle concentrations to the ambient air in all measured urban Indian households. We consider that poor ventilation systems hinder particles from being removed quickly from the air during cooking processes. In some areas, outdoor combustion processes also increased indoor particle levels. It is possible to reduce exposure risk and health effects caused by poor indoor air in urban Indian households by improving indoor ventilation, for example, by increasing the fan speed and opening windows and doors especially during cooking processes. Reducing the penetration of outdoor particles in the evenings is possible by isolating the gaps in the windows and doors. In some areas, installing of proper windows would decrease the indoor particle levels in the evenings.

These two studies have provided for the first time initial information on indoor number concentration levels, exposure patterns and correlations with CO and aerosol mass concentration in urban Indian households. However, much more data are required on aerosol exposures characterized by metrics other than mass concentration in India if associations between exposure and ill health are to be further understood and health risks reduced.

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