ABSTRACT

Particulate pollution has emerged as a serious environmental health concern in Pakistan. The use of biomass fuels in traditional stoves produces high levels of indoor air pollutants. In Pakistan, 94% of rural and 58% of urban households depend on biomass fuel. This study investigates variations in indoor/outdoor concentrations of particulate matter during various activities for three different micro-environments in Pakistan. At a rural site, the average indoor/outdoor ratios for PM_{10}, $PM_{2.5}$ and PM_1, in kitchens using biomass fuels were 3.80, 4.36 and 4.11, respectively. A large variation was recorded in the mass concentration of particulate matter during cooking with concentrations in the range 4,000 to 8,555µg/m^3. In a living room at rural site, the average indoor/outdoor ratios for PM_{10}, $PM_{2.5}$ and PM_1 were 1.74, 2.49 and 3.01, respectively. At the urban site, the average indoor/outdoor ratios for same size fractions were 1.71, 2.88 and 3.47, respectively. Cooking, cleaning and smoking were identified as principal contributors to the high indoor levels of particulate matter. This study showed considerably high concentrations of particulate matter, particularly in kitchens using biomass fuels, as compared to living areas. Thus women and children face the greatest exposure due to the amount of time they spend in the kitchen.

Practical Implications

In the developing world, particulate air pollution both indoor and outdoor is a substantial health hazard to public. The very high concentrations of particulate matter in both rural and urban sites, particularly in kitchens using biomass fuels emphasize the severity of this issue in Pakistan The women and children are extensively at risk due to amount of time spent in kitchens. This state of affairs calls for a large scale intervention to reduce the exposure to indoor air pollution.

INTRODUCTION

The sources and levels of air pollutants in developing countries are significantly different from those in the developed world. In the developing countries, population explosion along with widespread industrialization coupled with urbanization has resulted in dense urban centres with poor air quality. The use of biomass fuels in rural areas subjects the population to high indoor air pollution. Worldwide, more than 3 billion people rely on solid fuels, including biofuels, for their energy needs (WHO, 2007a). Due to incomplete combustion the use of biomass fuels in traditional stoves produces high levels of indoor air pollutants. This smoke contains a range of health deteriorating substances that, at varying concentrations, can pose a serious threat to human health. Indoor air pollution is responsible for more than 1.6 million annual deaths and 2.7 % of global burden of diseases (WHO, 2006).

Hence exposure to indoor air pollution from the combustion of biofuels (wood, dung, crop residues, charcoal) is a significant public health hazard predominately affecting the poor in both rural and urban communities in developing countries. For example there is strong evidence that smoke from biofuels can cause acute lower respiratory infection in childhood (WHO, 2006; Smith et al. 2000; Ezatti and Kammnen, 2001). A recent report on national burden of diseases from indoor air pollution by World Health Organization (2007a) confirms the linkage between indoor air pollution due to solid fuels and different diseases, including acute and chronic respiratory diseases, tuberculosis, asthma, and cardiovascular disease and prenatal health outcomes. In most of cases indoor air pollution disproportionately affects women and children who spend most time near the domestic hearth.

Air Pollution and Pakistan

The Government of Pakistan commenced a National Environment Action Plan in 2001 with the support of the United Nations Environment Programme. Work has concentrated primarily on developing policies. The Pakistan Clean Air Programme has identified

vehicular emissions, industrial emissions, burning of solid waste and natural dust as major sources of urban air pollutants in Pakistan. However, despite recognizing the severity of air pollution little work has been undertaken on integrated air quality assessment and management systems (Qadir, 2002). Only scattered information is air pollution measurements. The State of the Environment Report (Pak-EPA, 2005) by the Pakistan Environmental Protection Agency identified degradation of ambient air quality as a major environmental concern. This report recognized industrial pollution, suspended particulates, indoor air pollution, and increasing traffic trends as key sources affecting ambient air quality in the country. It is note worthy that, though indoor air pollution was mentioned as key source of affecting ambient air quality yet it was discussed with reference to excessive biomass fuel use and high indoor air pollution in rural areas. Currently there are no ambient air quality standards. Recently Ghauri et al. (2007) presented the results of a year long base line air quality study in Pakistan during 2003-2004. The highest concentrations of O_3 (50 ppb), SO_2 (52.5 ppb) and NO_X (60.75 ppb) occurred in Lahore while peak CO (14 ppm) levels were reported for Quetta. Overall, the concentrations of O_3, SO_2 and NO_X were within the limits of the US-EPA standards except 1 hour average of CO was exceeded at Lahore, Karachi and Quetta. However, PM_{10} levels were particularly high and maximum levels at Lahore, Peshawar, Quetta, Karachi, Islamabad and Rawalpindi were 368 $\mu g/m^3$, 350 $\mu g/m^3$, 331 $\mu g/m^3$, 302 $\mu g/m^3$, 280 $\mu g/m^3$ and 276 $\mu g/m^3$, respectively. The mean PM_{10} levels exceeded the US EPA standard limit of 150 $\mu g/m^3$. The concentration of hydrocarbons (Methane) were ranged from 0.1 – 2.8 ppm. While, the concentrations of nonmethane hydrocarbons ranged from 0.1 to 3.2 ppm.

Indoor air pollution

Pakistan, with almost 70% of population living in rural areas, uses wood, dung, crop residue or natural gas as a fuel for cooking and heating. These biomass fuels produce high levels of indoor air pollution and pose a serious threat to health of inhabitants. This situation is worse in cities where outdoor air pollution adds to that generated indoors. The use of biomass fuel in Pakistan is 86% with 54% using wood (Archar, 1993). According to the World Health Organization (2007b), in Pakistan indoor air pollution due to solid

fuel use is responsible for 4.6 % of national burden of disease. With regard to indoor air pollution in Pakistan, there is little published evidence. Recently, a study was undertaken on the correlation of eye and respiratory symptoms among women exposed to wood smoke emitted from indoor cooking and concluded that these are significantly associated with wood use (Siddiqui et al., 2005a). Another study showed an independent effect of indoor air pollution on birth weight (Siddiqui et al., 2005b). A study by Akhtar et al. (2007) in the rural area of Peshawar, Pakistan revealed a strong association of biofuel smoke exposure with chronic bronchitis in women who are involved in cooking with biomass fuels. Studies on indoor air pollution from solid fuels have been conducted in various developing countries in recent years e.g. Mexico (Zuk et el. 2007), Philippines (Saksena et al. 2007), China (Fischer and Koshland 2007; Mestl et al., 2007), Zimbabwe (Rumchev et al. 2007), Bangladesh (Dasgupta et al., 2006), India (Balakrishnan et al., 2002, 2004), Costa Rica (Park and Lee 2003), Bolivia (Albalak et al., 1999) and Kenya (Boleij et al., 1989). But no study on indoor particulate matter levels has been carried out in Pakistan yet. According to the WHO (2005) despite mounting evidence that biomass smoke exposure increases the risk of a range of diseases little intervention is being done in Pakistan. Hence there is a dire need to monitor levels of indoor air pollution in Pakistan. Most of these studies conducted in other developing countries reported mass concentrations in either PM_{10} or $PM_{2.5}$ size fraction from kitchens using biomass fuels. However, studies on indoor/outdoor correlation of PM_{10}, $PM_{2.5}$, PM_1 in kitchens and living rooms are very rare. Therefore, the present work was carried out to investigate the relationship between indoor/outdoor air quality and to assess the levels of indoor air pollution in rural and urban environments in Pakistan. The objectives were to:

i) monitor the mass concentration of particulate matter (PM_{10}, $PM_{2.5}$, PM_1) in different indoor and outdoor micro-environments in Pakistan.

ii) investigate the variation in indoor/outdoor particulate matter ratios.

iii) assess pollution levels during various indoor and outdoor activities.

These were achieved by a series of real time measurements of particulate matter by using two aerosol spectrometers. This work has, not only, provided a snapshot of the current situation of indoor air pollution but will provide baseline information to prioritise the future studies.

MATERIALS and METHODS

Sampling Sites

To investigate the indoor/ outdoor air quality in Pakistan sampling was carried out during November 2005 to February 2006. Sites were selected to reflect different households and fuels. During this time of the year the cooking in rural areas was carried out in closed space kitchens as compare to summer when usually it is conducted in outdoors. Air samples were collected from two rural sites (Site I - Chak NO.35/2.L and Site II - Bhaun) and an urban site (Lahore). At rural site I (Chak NO.35/2.L) the sampling was carried out in two different kitchens and one living room. Both kitchens used biomass fuel for cooking. The kitchens were detached from living rooms. At rural site II (Bhaun) and the urban site (Lahore) sampling was conducted in the living rooms. At these sites natural gas was used as a fuel. The majority of houses at site I were made of mud, grasses and bamboo. The courtyards of these were generally not tiled and devoid of any grass. The rural site II was in a semi urban area and there was a range of houses of different construction materials. Lahore, the urban site, is one the mega-cities of Pakistan. Here sampling was undertaken in one of the slums of the city. The ventilation in all the cases was through windows or doors and they remained open during the day and were closed during the night. All the sampling sites had electric supply for lighting, however in rural areas during electric break down use of kerosene lamps is a common practice. But during our study no kerosene lamps were used in any of the experimental space. The detailed description of the houses is shown in table 1.

Instrumentation

The mass concentration of particles (PM_{10}, $PM_{2.5}$, PM_1) was monitored using two GRIMM aerosol spectrometer: i) Model 1.108 ii) Model 1.101 (Grimm Aerosol Technik GmbH, Ainring, Germany). The GRIMM monitors have a sensitivity of 1 particle/litre with a reproducibility of ± 2%.

Both of the aerosol spectrometers work on the following same principal. An air sample is collected by a radial symmetry sampling head and is constantly drawn into an optical chamber. Each single particle is detected by light scattering at 90º with a high speed photodiode. An integrated pulse height analyzer then classifies the signal into different size ranges. Software allows the data to be viewed as counts /l or mass as µg/m^3. Both of these aerosol spectrometers were factory calibrated, prior to the sampling campaign. A gravimetric correlation was carried out with Stearin and an optical calibration cross reference was performed with spherical glass beads with a density of 2.8g/cm^3 and a refractive index of 1.36.

The model 1.108 can classify up to 15 size ranges and has a flow rate of 1.2 l/minute. The Model 1.101 reports only the mass fraction in 3 size distributions and operates at a flow rate of 0.60 l/minute. For the present study both of spectrometers were used for the report mass fraction in the environmental mode (PM_{10}, $PM_{2.5}$, PM_1). Both of these monitors were operated side by side in each experimental setting for 12 hours before the start of sampling and intercomparision between them revealed a variation of +/- 10 %.

The model 1.108 was always used for the indoor measurements and 1.101 for outdoors. The stoves in both the kitchens were in a hole in the ground of approximately 30 cm x 30 cm x 15 cm. There was no chimney installed above them. In kitchens the instrument was placed approximately 60 cm away from the stove, corresponding to the distance between the stove and the sitting position of the person cooking. The instrument was placed on table of height 0.8 m and 30 cm from the person cooking. In the living room in both the rural and urban sites the instruments were placed at height of 1 m above the ground and 1.5 m away from doors and windows. The sampling outdoors was carried out in

courtyard of the household approximately 2 m away from the living rooms. The monitor was placed at the height of 1 m. The sampling was carried out indoor and outdoor simultaneously. Both of these instruments were run continuously for a period of one week in each setting and data recorded every one minute. A temperature and humidity logger (Model RH-02, Pico Technologies Limited, UK) was used to record the indoor temperature and humidity in all the sampling sites. The data was collected with a sampling interval of 1 minute.

The activities of the inhabitants were documented during the sampling periods. 24-hr time–activity diaries for each experimental space were maintained throughout the sampling period. Activities in kitchen were divided into the following categories: cooking (e.g., preparation for cooking, lighting, and tending the fire), cleaning, and other (e.g., outside the floor sweeping, no activity). In the living room activities were socializing (e.g., smoking, conversing, watching television), cleaning, sleeping at night time and floor sweeping outdoors. Periods of no activity were also identified by the analysis of diaries. These periods were defined as when there was no known activity within a radius of 20 meters of experimental space. The data was further analysed hourly to investigate the effect of various activities on particulate levels and 24 hourly averages were also calculated. The effect of smoking and cleaning in living room was documented. A regression analysis was carried out to investigate correlations between indoor/outdoor PM_{10}, $PM_{2.5}$ and PM_1.

Results and Discussion

Temperature and humidity

The sampling was carried out during the winter time. The days were usually sunny with clear skies. However ambient temperature fell significantly during the night. The 24 hour average indoor temperature and relative humidity in the kitchen at rural site I was 17°C (range: 8 - 26°C) and 48% (range: 36 - 58%) respectively. While in the living rooms at the rural site the average indoor temperature and humidity was 14°C (range: 10 - 24°C) and 52% (39 - 65%), respectively. The indoor temperature and humidity at the urban site

was 19°C (range: 10 - 21°C) and 39% (range: 29 - 54%) respectively. The average ambient temperature at rural sites was 16°C with a maximum of 26°C and minimum of 4°C. The temperature at the urban site ranged from 5 - 28°C with an average of 19°C. The ambient relative humidity at both urban and rural sites was in the range 39 - 77% with an average of 65%. The highest relative humidity was recorded during the early hours of the morning. These outdoor measurements were taken in courtyards at a height of 1 m and 2 m away from buildings.

Mass Concentration of Particulate Matter in a Kitchen Using Solid Mass Fuels (Rural)

The 24 hour average indoor concentration of PM_{10}, $PM_{2.5}$ and PM_1 were1581 µg/m^3, 1169 µg/m^3 and 913 µg/m^3, whereas, for the same size fraction the outdoor levels were 310 µg/m^3, 182 µg/m^3 and 165 µg/m^3, respectively (Table 2). The hourly indoor /outdoor (I/O) ratios for PM_{10} were in the range 0.61 to 26.5 with a 24 hour average of 3.80. Similarly, the average indoor/outdoor ratios of $PM_{2.5}$ and PM_1 were 4.36 and 4.11 with a range of 0.96-33.93 and 0.91-31.59, respectively (Table 2). The highest indoor/outdoor ratios were obtained during the cooking hours whilst periods with no cooking or no activity the hourly ratios were close to unity for both $PM_{2.5}$(0.96) and PM_1(0.91) (Table 2). A sharp fall in I/O ratio of PM_{10} (0.60) was recorded during cleaning in the courtyard. These results suggest that although the particulate levels were substantially high during cooking they fell sharply once it had stopped and indoor levels were approximately those outdoors. However, this behaviour largely depends on the ventilation rates of the space. The kitchens in the present study had no chimneys but they had a window and door open during cooking and afterwards. Furthermore, over a period of week a periodic rise and fall in levels of PM_{10}, $PM_{2.5}$ and PM_1 was seen corresponding to three meals cooked over the day. This provides evidence of the episodic exposure to high levels of particulate matter for people who spend time close to the fireplace. With reference to coarse fraction, on average higher $PM_{10} - PM_{2.5}$(311 µg/m^3) was obtained in indoors than outdoors (127 µg/m^3). The maximum coarse fraction was recorded during cleaning events. However, it has been noted that although cooking contributes more to the fine fraction, during the first hour of cooking a moderate increase in $PM_{10} - PM_{2.5}$ was observed. This probably

reflects the process of resupesion due to the physical movement of people involved in cooking during the period of setting the fire in the stove. This period varies and is largely dependant upon the dryness of biomass fuel and size/design of the stove.

Effect of Cooking and Cleaning

In general, a large variation was observed in mass concentration of particulate matter during cooking with concentrations in the range of 4,000 $\mu g/m^3$ – 8,555 $\mu g/m^3$ (Figure 1). A study conducted in rural Tamil Nadu, India by Parikh et al. (2001) reported PM_{10} concentration in the range from 500 – 2000 $\mu g/m^3$ during a two hour cooking period using biofuel. They suggested that the variation was the result of changing ventilation rates and non-uniform combustion rates.

A large variation in mass concentration of particulate matter has been recorded during a 24-h cycle within the kitchens. Such variations were primarily due to the contribution from biofuel smoke inside the kitchen or cleaning of the courtyard outside. These findings are in agreement with Park and Lee (2003), who reported on particle exposure and size distributions from wood burning stoves in Costa Rica. They pointed out that particulate levels increased rapidly during cooking and decreased quickly after cooking. In their study the maximum peak particulate levels ranged from 310 to 8170 $\mu g/m^3$ for $PM_{2.5}$ and from 500 to 18,900 $\mu g/m^3$ for PM_{10}.

The cooking time in our sampling kitchens was ranged from 2-3 hours and cooking and cleaning was identified as principal activities. Although the sampling was carried out in both kitchens separately for a period of one week the levels of PM_{10}, $PM_{2.5}$ and PM_1 were averaged over all the cooking and cleaning events over a period of seven days, because the levels in both the sampling kitchens showed the same trend. Over a period of one week, the daily levels of PM_{10}, $PM_{2.5}$ and PM_1 during cooking ranged from 1991 $\mu g/m^3$ to 7881 $\mu g/m^3$, 1531 $\mu g/m^3$ to 2664 $\mu g/m^3$ and 1430 $\mu g/m^3$ to 2396 $\mu g/m^3$, respectively. The concentration of PM_{10} – $PM_{2.5}$ was low (460 $\mu g/m^3$ to1202 $\mu g/m^3$) as cooking with biomass fuel had the biggest contribution in the fine fraction rather than the coarse. On average, in both kitchens, during cooking episodes the concentrations of PM_{10}, $PM_{2.5}$ and

PM_1 were approximately 4,000 µg/m^3, 3000 µg/m^3 and 2500 µg/m^3 with a background value of 300 µg/m^3 , 270 µg/m^3 and 240 µg/m^3 for PM_{10}, PM $_{2.5}$ and PM_1 (Figure 2). The back ground value was averaged from a period of no activity as during these periods the levels were fairly stable. Recently Dasgupta et al. (2006) conducted a study on indoor air quality in Bangladesh and reported that PM_{10}, over 24hr cycle, in wood using households, varied from 68 to 4,864 µg/m^3. It has been seen that in kitchens it can take up to an hour for the indoor air to reasonably clear after cooking. Generally we observed a wide variation in concentration of particulate matter among different kitchens and even within the same kitchen during different episodes of cooking. The variation primarily depended on the quality (dryness) of biomass fuel used, duration of cooking, degree of incomplete combustion and ventilation. Ezzati and Kammen (2002) mentioned that a typical 24-hr average concentration of PM_{10} in homes using biofuels may range from 200 to 5,000 µg/m^3 or more throughout the year, depending on the type of fuel, stove, and housing characteristics and significant temporal and spatial variations may occur within a house.

On the other hand, during cleaning the daily levels of PM_{10}, PM $_{2.5}$ and PM_1 ranged from 1507 µg/m^3 to 3724 µg/m^3, 201 µg/m^3 to 1224 µg/m^3 and 139 µg/m^3 to 1557 µg/m^3, respectively. The levels of PM_{10}- PM $_{2.5}$ were in the range of 546 to3,004 µg/m^3. During cleaning events a rise in the coarse fraction was recorded. However, an increase in PM_1 was observed when cleaning was carried out shortly after cooking and space was not completely cleared of smoke. Furthermore, the kitchen floor was not tiled but compacted bare earth. Hence, cleaning resulted in the generation of high levels of coarse dust. The higher PM_{10} – PM $_{2.5}$ levels during cleaning as compared to cooking reflect this behaviour (Figure 3). The average levels of PM_{10}, $PM_{2.5}$ and PM_1 during different cleaning events in the kitchens were approximately, 2714 µg/m^3, 885 µg/m^3 and 725 µg/m^3, respectively. It should be noted that although cleaning contributed more to the coarse size fraction the levels of PM_1 were almost double those for periods of no activity. Most of the cleaning events were carried out after breakfast and these higher levels were probably due to residual smoke in the kitchen space. Time –activities diaries illustrated the presence of only women who cook or who stay close to stove and children under the

age of 5 years during the cooking periods. This is very likely that women and young children were receiving the highest exposure to indoor particulate matter during cooking while men were rarely in the kitchen during the cooking periods.

Mass Concentration of Particulate Matter in Living Room (Rural)

The average indoor concentrations for PM_{10}, $PM_{2.5}$ and PM_1 were 953 µg/m^3, 603 µg/m^3 and 548 µg/m^3, respectively. While the average outdoor concentrations for the same size fractions were 2,838 µg/m^3, 413 µg/m^3 and 203 µg/m^3, respectively (Table 3). Hourly average concentrations exhibited a wide variation with peak PM_{10} levels, up to 21,673 µg/m^3, occurring in the floor early in the morning as a result of sweeping.

In general, during most of the day indoor concentrations were higher than those outdoors, except in the morning (Figure 4). Furthermore, the indoor/outdoor ratio confirms this behaviour and the 24 hour average I/O ratios for PM_{10}, $PM_{2.5}$ and PM_1 were 1.74, 2.49 and 3.01, respectively (Table 3). The results suggest an indoor source of fine particulates. The only known source was smoking. During smoking a sharp rise in fine particulates was observed and the I/O ratio for PM_1 rose to a maximum of 12.95. Increases were also observed for $PM_{2.5}$ (2.49) and PM_{10} (5.77). A study by Jones et al. (2000), showed that mean daily I/O ratios of PM_{10} in smoking homes of rural Oxfordshire were greater than unity (2.7 ± 6.7). Monn et al. (1997) reported that indoor smoking had the highest influence on I/O ratios during an investigation of 17 houses in Zurich, Switzerland. They reported that in houses with smokers daily mean I/O ratio of PM_{10} was 1.84 and 2.07. Indoor/Outdoor ratios for $PM_{2.5}$, measured as part of the EXPOLIS study, showed that the 48 hours ratios, in smoking houses, was invariably greater than unity and ranged from 1.41 to 2.09 for various European cities (Götschi, et al. 2002). The I/O ratios of $PM_{2.5}$ and PM_1 in this study were relatively higher than any other reported studies. This was probably due to the differences in environment, air exchange rates and, more importantly, social behaviour. Sampling was undertaken in a typical Pakistani rural living room, shared by 4 to 8 people over a 24 hour period, with highest occupancy in the evening during traditional social gatherings. Analysis of PM_{10} - $PM_{2.5}$ showed that, on average, outdoors had the highest coarse concentrations (2,425 µg/m^3) in comparison to indoors

(350 µg/m^3). (Table 3). These high outdoor values were most probably due the sweeping of the courtyard as it was devoid of any grass/vegetation. A similar trend was seen indoors, with high levels of coarse particulate matter during cleaning. Although the average indoor PM_{10} – $PM_{2.5}$ values were much lower than outdoors and most of the indoor activities contributed to the fine fraction. However during the evening social gatherings the levels of the coarse size fraction were slightly higher with a range of approximately 500 to 1000 µg/m^3. The highest increase was recorded during the first hour of the gathering and was probably due to extensive physical movements by indoor occupants resulting in the resuspesion of settled dust from indoor surfaces. Moreover, the standard deviation values of outdoor PM_{10} – $PM_{2.5}$ were substantially higher than indoor PM_{10} – $PM_{2.5}$ reflecting that there was less variation in the indoor environment.

Effect of Smoking and Cleaning

The mass concentration of particulate matter as a result of cigarette smoking revealed a sizeable increase in PM_1 and corresponding increases in $PM_{2.5}$ and PM_{10}. During smoking PM_{10} concentrations ranged from 2,100 µg/m^3 to 2,700µg/m^3, while background values indoors were approximately 400 µg/m^3 (Figure 5). Indoor background levels were always higher than those outdoors except during the early morning when sweeping was taking place. The high indoor background concentrations of particulate matter, even when no apparent indoor source was in apparent, could be due to residual tobacco smoke; the majority of the particulate matter is in the PM_1 fraction. A comparison of these results with a non-smoking living room supports this. The mass concentration of particulates matter in a non smoking living room of roughly the same volume, although of different construction material, doesn't exceed 110µg/m^3 (Figure 6), even during cleaning. Background values were also low (40 µg/m^3) in comparison with the smoker's living room (400 µg/m^3). A clear contribution to PM_{10} from outdoors is evident, due to the window opening towards the street (Figure 6). Sweeping results in a large rise of PM_{10} both indoors and outdoors (Figure 4). However outdoors the concentration was approximately 10 times higher than that indoors (21,000 µg/m^3 vs 2,300 µg/m^3). Due to the dry conditions the court yard was extremely dusty and no vegetation was present.

Although these values show short term increases in particle concentration as a result of smoking and sweeping they are of importance with regard to human exposure.

Mass concentration of particulate matter in a living room (urban)

In Lahore the mass concentration of particulate matter fluctuated due to the indoor activities (Figure 7) of the inhabitants and outdoor sources. The 24 hour average indoor mass concentrations of PM_{10}, $PM_{2.5}$ and PM_1 were 533 µg/m^3, 402 µg/m^3 and 362 µg/m^3, respectively. On the other hand outdoor 24 hour mass concentrations of PM_{10}, $PM_{2.5}$ and PM_1 were 308 µg/m^3, 142 µg/m^3, 109 µg/m^3, respectively (Table 4). The room (student accommodation shared by three people) was located in a city slum with heavy traffic nearby. The indoor concentrations were generally higher than those outdoors. The room remained unoccupied for 6 – 8 hours during the day and the occupants were smokers.

The indoor/outdoor (I/O) ratio for PM_{10} varied from less than 1 to 3.7 with those below occurring when the room was unoccupied. Peak indoor concentrations $PM_{2.5}$ and PM_1 were higher that those measured outdoors again highlighting the importance of cigarette smoke. The I/O ratios for these two size fractions ranged from below 1 up to 8.5 (Table 4). A large fluctuation was detected in the living room due to different activities (e.g. cleaning , smoking, walking, dressing). During the afternoon indoor concentrations have fallen close to those outdoors due to the room being unoccupied. Although the lowest indoor concentrations were during the afternoon the maximum I/O ratios were in early hours of morning when the outdoor levels were far lower than those indoors. In a study by Monn et al. (1997) the mean I/O ratio for PM_{10} in 17 houses with human activity was 1.40. The average levels of PM_{10} – $PM_{2.5}$ indoors was 128 µg/m^3, while the average outdoor levels were slightly higher (166 µg/m^3) (Table 4). The highest indoor coarse fraction was correlated with indoor occupancy. The indoor social activities (smoking, watching television) in this room continued late into the night and these might have resulted in the resuspension of particulate matter. In addition levels of PM_{10}, $PM_{2.5}$ and

PM_1 were high throughout night. This was due to different sleeping schedules of the occupants and closed doors and windows resulting in reduced ventilation.

Comparison of Rural and Urban Living Rooms

The 24 hour average indoor mass concentration of PM_{10}, $PM_{2.5}$ and PM_1 in the rural living room was 953µg/m^3, 603 µg/m^3 and 548µg/m^3 in comparison with 533 µg/m^3, 402 µg/m^3 and 362 µg/m^3, respectively, in the urban living room. On the other hand outdoor 24 hour mass concentrations of PM_{10}, $PM_{2.5}$ and PM_1 in the rural area were 2838 µg/m^3, 413 µg/m^3, 210 µg/m^3 as compared to 308 µg/m^3, 142 µg/m^3, 109 µg/m^3 respectively at the urban site. This comparison revealed that both indoor and outdoor levels of particulate matter were higher in rural areas. Outdoor concentrations in this study reflect the concentration 5 metres away from indoor settings. The high levels of particulate matter in the rural living room are primarily due to indoor smoking. At Bhaun (Rural Site II), the living room with no smokers experienced a maximum mass concentration far lower (110µg/m^3) than at both Chak NO.35/2.L (Rural Site I) and Lahore (Urban site). With reference to outdoor levels in rural areas agricultural practices, sweeping and biomass burning are the principal contributors, while at the urban residential site the major source was automobile exhaust. With regard to $PM_{10} - PM_{2.5}$ levels, higher values were obtained at the rural sites, both indoors and outdoors, than at the urban site. This may be the result of different physical characteristics of micro environments and activities of the occupants.

Correlation Between Indoor and Outdoor PM_{10}, $PM_{2.5}$ and PM_1

In order to evaluate the effect of outdoor concentrations on those indoors, a regression analysis was carried out taking the indoor concentration as dependent variable and outdoor as an explanatory variable. The following regression equation was considered.

$$(C)_I = \acute{\alpha} + \beta(C)_O + \epsilon$$

Where, C is the concentration of PM in different size fractions, I and O refer to indoor and outdoor concentration, ά is a constant, β is a regression coefficient and ϵ is the random error with zero mean.

The t and p values for PM_{10}, $PM_{2.5}$ and PM_1 at rural living room revealed that we cannot reject the null hypothesis that $\beta = 0$ (Table 5). Furthermore, the goodness of fit (R^2) values for PM_{10}, $PM_{2.5}$ and PM_1 were 0.015, 0.027 and 0.083, which suggest that only 1.5%, 2.7% and 8.3% variation in the indoor concentration of PM_{10}, $PM_{2.5}$ and PM_1, can be attributed to outdoor concentration respectively (Table 5) The very low values of R^2 revealed the independent sources of particulate matter in the indoor environment. In the case of the living room, indoor smoking and cleaning were identified as the principal contributors towards PM. On the other hand at urban site, the t and p values for all size fractions suggest that the null hypothesis can be rejected and β was significant at < 0.01%. Hence there was a significant impact of outdoor PM on that indoor; results indicate that 61%, 63% and 59% variation in indoor levels of PM_{10}, $PM_{2.5}$ and PM_1 can be due that outdoors (Table 4). A study in California (Geller et al., 2002) which focused on PM_{10} and $PM_{2.5}$ showed a weak correlation (R^2=0.35) between outdoors and indoor coarse particles (2.5 - 10 µm). They concluded that the outdoor concentration accounted for only 37% of the variation in indoor concentrations. In the present study, a significant correlation was found at the urban site living room between indoor/outdoor PM_{10}, $PM_{2.5}$ and PM_1 while none was found for the rural site living room. The duration of occupancy of the living room was different at both the sites. At rural sites there were various activities during the most of the day, whilst the urban living room was mostly unoccupied during the daytime. Hence, it is very likely that over a period of 24 hours, at the urban site the indoor levels of particulate matter were approximately same as those outdoors. The higher goodness of fit (R^2) values for the urban living room could be due to different occupancy duration. Furthermore, the higher average indoor values of PM_{10} (953 µg/m^3), $PM_{2.5}$ (603 µg/m^3) and PM_1 (203 µg/m^3) in the rural living room than in the urban living room (PM_{10} (533 µg/m^3), $PM_{2.5}$ (402 µg/m^3) and PM_1 (109 µg/m^3)) provides evidence of greater indoor occupancy. It was observed that indoor occupancy was mostly related to a particulate matter generating activity.

In the kitchens at the rural site regression analysis of hourly indoor and outdoor averages over a period of one week showed different patterns than in rural living rooms. The t and p values for PM_{10}, $PM_{2.5}$ and PM_1 suggest that the null hypothesis can be rejected and β was significant at < 0.05%. (Table 6) This implies that there was a significant impact of outdoor particulate matter on indoor levels. However there was a relatively low goodness of fit value but is was still significantly higher than for the living room. The goodness of fit (R^2) values for kitchen indicate that 28%, 27% and 22% of the variation in indoor levels of PM_{10}, $PM_{2.5}$ and PM_1 can be due to that outdoors (Table 6). This phenomenon is most probably due to different occupancy/activity patterns. In the investigated kitchen three meals were cooked during the day and after cooking it was generally unoccupied. The total time for these meals was 6 – 8 hours/ day. Although during the cooking periods the levels were substantially high they fell sharply afterwards. This gives an indication that apart from cooking and cleaning for the remainder of the day indoor levels corresponded to those outdoors.

Conclusion

In summary, the results of this study showed a large variation in the I/O ratio for particulate matter at rural and urban sites in Pakistan. The levels of particulate matter in both rural and urban areas were higher than any established standard. In rural areas the use of biomass fuel was a principal contributor for high concentrations of particulate matter in kitchens and smoking had a major share in the deterioration of living room air quality. Overall indoor concentrations were higher than those outdoors, in both rural and urban areas. The present study shows considerably high concentrations of particulate matter, particularly in the kitchen using biomass fuel as compared to the living areas. Thus women and children are exposed the most due to amount of time they spend in the kitchen. These concentrations were many times in excess of EU, US EPA and WHO standards/guidelines. This situation of indoor air quality warrants the need to take serious steps to improve it. An intervention based on needs of the users at the community levels is urgently required to cut down the exposure and to improve public health. A recent report by the WHO (2007) strongly supports the case for giving high priority to measures to control indoor air pollution. Although the present investigation has studied only a few

households yet it is the first to measure indoor/outdoor levels of PM_{10}, $PM_{2.5}$ and PM_1 in Pakistan. A more detailed study with more households using different fuels and focused on all parameters affecting indoor air quality should be carried out.

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Table 1. General Description of Sites

Site	Area	Experimental space	Age of the House	Volume (m^3)	Ventilation	Fuel used/ activity
Rural Site- I (Chak NO. 35/2.L.)	Rural, residential, lots of agricultural land, low traffic density, mud buildings, large number of livestock in most houses.. No paved streets. Lighting with electricity.	Living room (combined, used by 3-7 persons)	20 yrs	72	Window opening (one)	None/ normal household activities, smoking
		Kitchen –I Floor: mud plaster Courtyard: untilled, no grass	4 yrs	20	Door/ window opening	Dung and crop residues/ cooking, cleaning
		Kitchen - II Floor: mud plaster Courtyard: untilled, no grass	2 yrs	20	Door /window opening	Dung and crop residues/ cooking, cleaning
Rural Site- II (Bhaun)	Semi urban, near road, low traffic density, paved streets, mud, concrete and iron shed buildings. Lighting with electricity.	Living room Floor: concrete Courtyard: untilled, no grass	16 yrs	60	Window/ door opening	None/ sleeping, reading and cleaning.
Urban Site (Lahore, Ichra)	Residential, densely populated, close to road, no greenery, within the shopping market.	Living room(carpeted) Courtyard: concrete floor	40 yrs	50	Window (two)	None/ student life, smoking, cleaning

Table 2. Summary of indoor/outdoor mass concentration of particulate matter in kitchens at rural site I

	Indoor PM_{10} ($\mu g/m^3$)	Outdoor PM_{10} ($\mu g/m^3$)	I/O PM_{10}	Indoor $PM_{2.5}$ ($\mu g/m^3$)	Outdoor $PM_{2.5}$ ($\mu g/m^3$)	I/O $PM_{2.5}$	Indoor PM1 ($\mu g/m3$)	Outdoor PM_1 ($\mu g/m^3$)	I/O PM_1	Outdoor PM_{10}-$PM_{2.5}$ ($\mu g/m^3$)	Indoor PM_{10}-$PM_{2.5}$ ($\mu g/m^3$)
Ave.	1581	310	3.80	1169	182	4.37	913	165	4.11	127	311
Max	8555	712	26.52	5953	381	33.93	3449	354	31.59	556	993
Min	141	67	0.62	23	42	0.96	13	36	0.92	22	26
St dev.	2003	223	6.10	1489	132	7.80	992	124	7.18	147	328

Table 3. Summary of indoor/outdoor mass concentration of particulate matter in living room at rural site

	Indoor PM_{10} ($\mu g/m^3$)	Outdoor PM_{10} ($\mu g/m^3$)	I/O PM_{10}	Indoor $PM_{2.5}$ ($\mu g/m^3$)	Outdoor $PM_{2.5}$ ($\mu g/m^3$)	I/O $PM_{2.5}$	Indoor PM1 ($\mu g/m3$)	Outdoor PM_1 ($\mu g/m^3$)	I/O PM_1	Outdoor PM_{10}-$PM_{2.5}$ ($\mu g/m^3$)	Indoor PM_{10}-$PM_{2.5}$ ($\mu g/m^3$)
Ave.	953	2838	1.74	603	413	2.49	548	203	3.01	2425	350
Max	2750	21673	5.77	2213	2087	10.80	2095	523	12.95	19587	2028
Min	288	210	0.03	170	89	0.35	148	71	1.28	94	29
St dev.	641	5193	1.51	421	495	2.07	400	117	2.31	4704	450

Table 4. Summary of indoor/outdoor mass concentration of particulate matter in living room at the urban site

	Indoor PM_{10} ($\mu g/m^3$)	Outdoor PM_{10} ($\mu g/m^3$)	I/O PM_{10}	Indoor $PM_{2.5}$ ($\mu g/m^3$)	Outdoor $PM_{2.5}$ ($\mu g/m^3$)	I/O $PM_{2.5}$	Indoor PM1 ($\mu g/m3$)	Outdoor PM_1 ($\mu g/m^3$)	I/O PM_1	Outdoor PM_{10}-$PM_{2.5}$ ($\mu g/m^3$)	Indoor PM_{10}-$PM_{2.5}$ ($\mu g/m^3$)
Ave	533	308	1.71	402	142	2.88	362	109	3.47	166	128
Max	1265	678	3.71	1030	333	6.70	940	281	8.37	422	253
Min	135	140	0.63	77	53	0.57	66	35	0.59	72	46
Std.Dev.	300	139	0.59	267	81	1.13	249	67	1.48	81	51

Table 5. Regression values for indoor PM_{10}, $PM_{2.5}$ and PM_1 on their corresponding outdoor concentrations in a rural and urban living room

	Rural					Urban				
PM Size	R^2	P- value	t- value	ά - value	β - value	R^2	P- value	t- value	ά - value	β - value
PM_{10}	0.015	0.576	-.568	996.42	-.0152012	0.61	<0.01	8.58	8.19	1.70
$PM_{2.5}$	0.027	0.45	.763	545.61	0.1399001	0.63	<0.01	8.97	29.52	2.61
PM_1	0.083	0.183	1.378	348.53	0.9846	0.59	<0.01	8.26	49.81	2.87

Table 6. Regression values for indoor PM_{10}, $PM_{2.5}$ and PM_1 on their corresponding outdoor concentrations in rural kitchens.

PM Size	R^2	P-value	t- value	ά - value	β - value
PM_{10}	0.30	<0.05	2.52	309	3.28
$PM_{2.5}$	0.30	<0.05	2.47	153	4.72
PM_1	0.20	<0.05	2.17	224	3.58

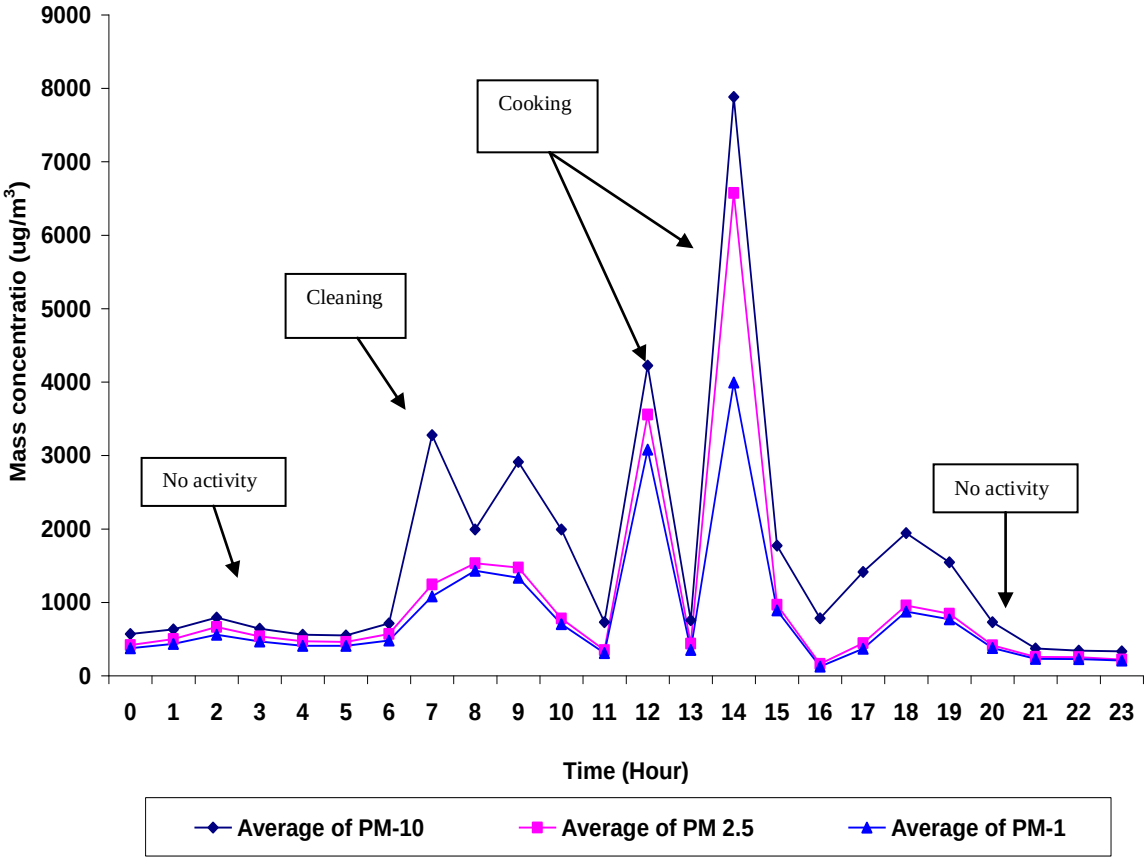

Figure 1. Representative hourly averages of mass concentration of PM_{10}, $PM_{2.5}$ and PM_1 in a kitchen using solid mass fuel at rural site in Pakistan on 27-12-05.

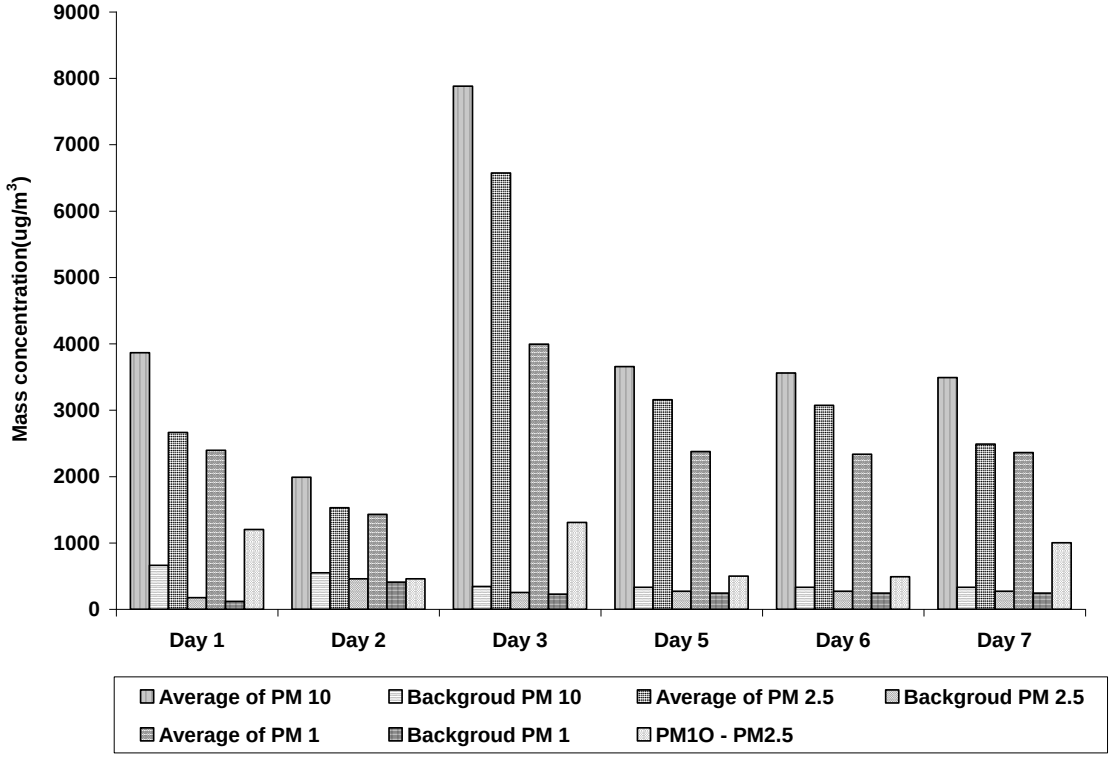

Figure 2. Average mass concentration of PM_{10}, $PM_{2.5}$ and PM_1 during daily cooking periods with solid mass fuel at rural site in Pakistan

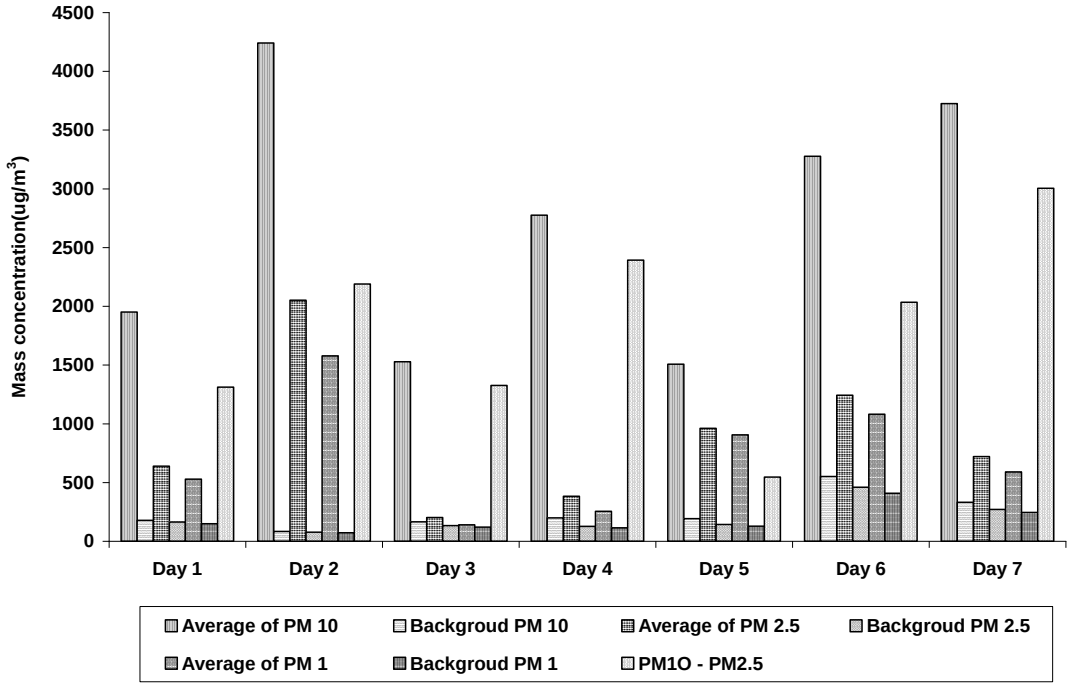

Figure 3. Average mass concentration of PM_{10}, $PM_{2.5}$ and PM_1 during daily cleaning periods in kitchens at rural site in Pakistan

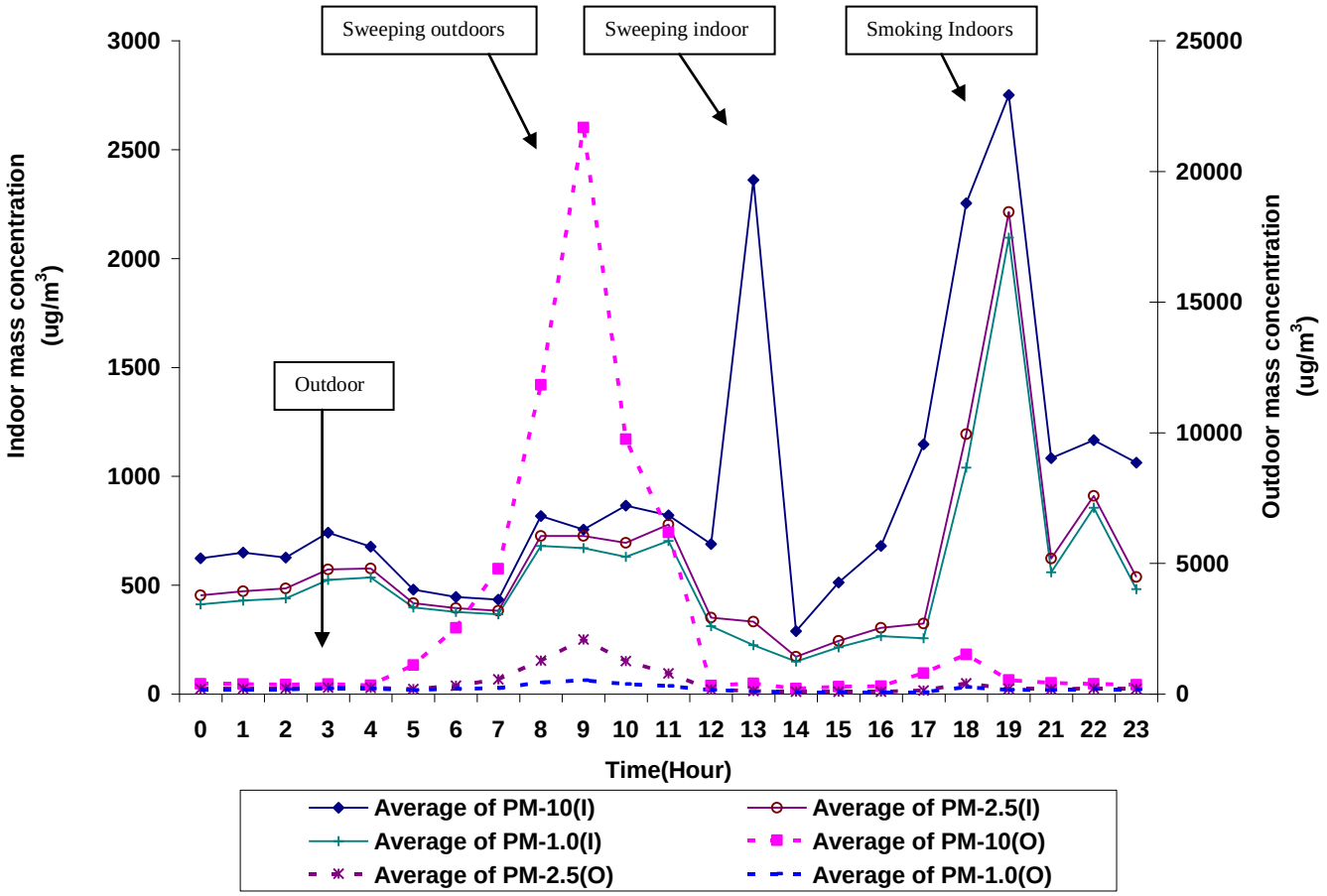

Figure 4. Representative hourly average of indoor and outdoor mass concentration for PM_{10}, $PM_{2.5}$ and PM_1 in a smoking living room at a rural site in Pakistan on 22 – 12 – 2005. I = Indoors; O = Outdoor

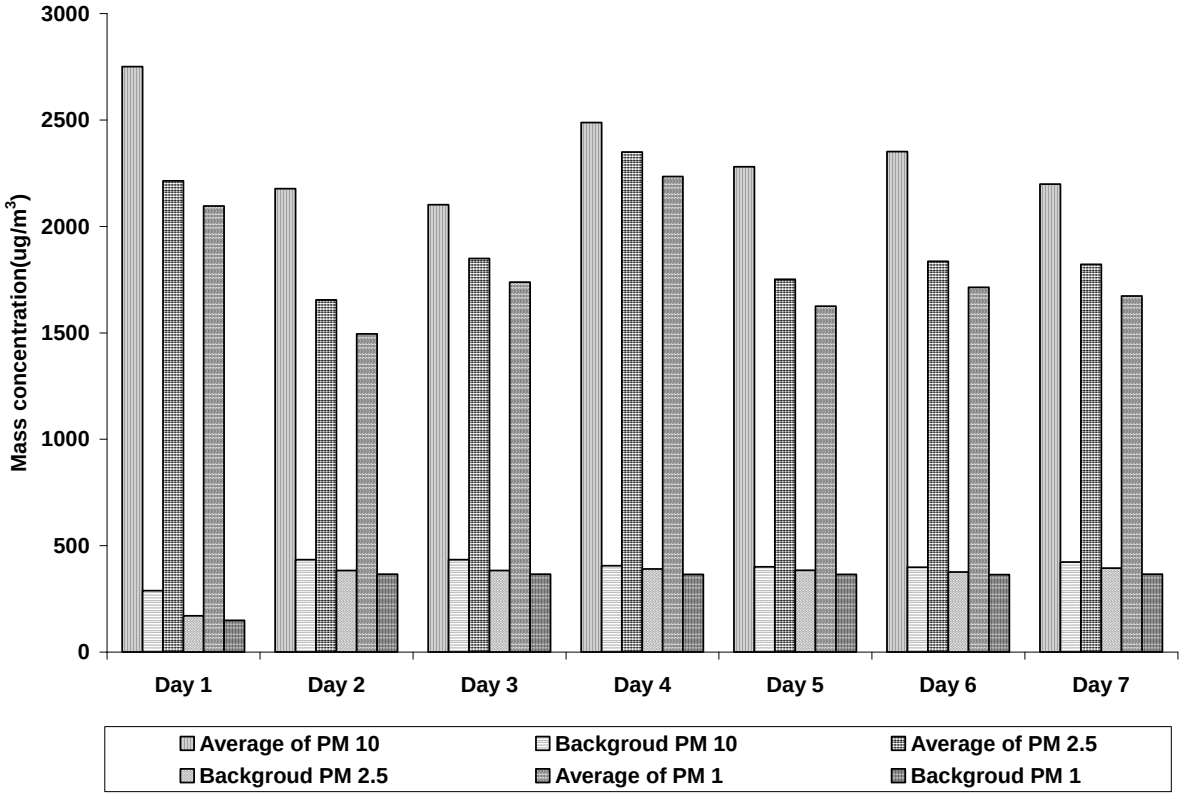

Figure 5. Average mass concentration of PM_{10}, $PM_{2.5}$ and PM_1 during various daily smoking periods in a living room at the rural site in Pakistan.

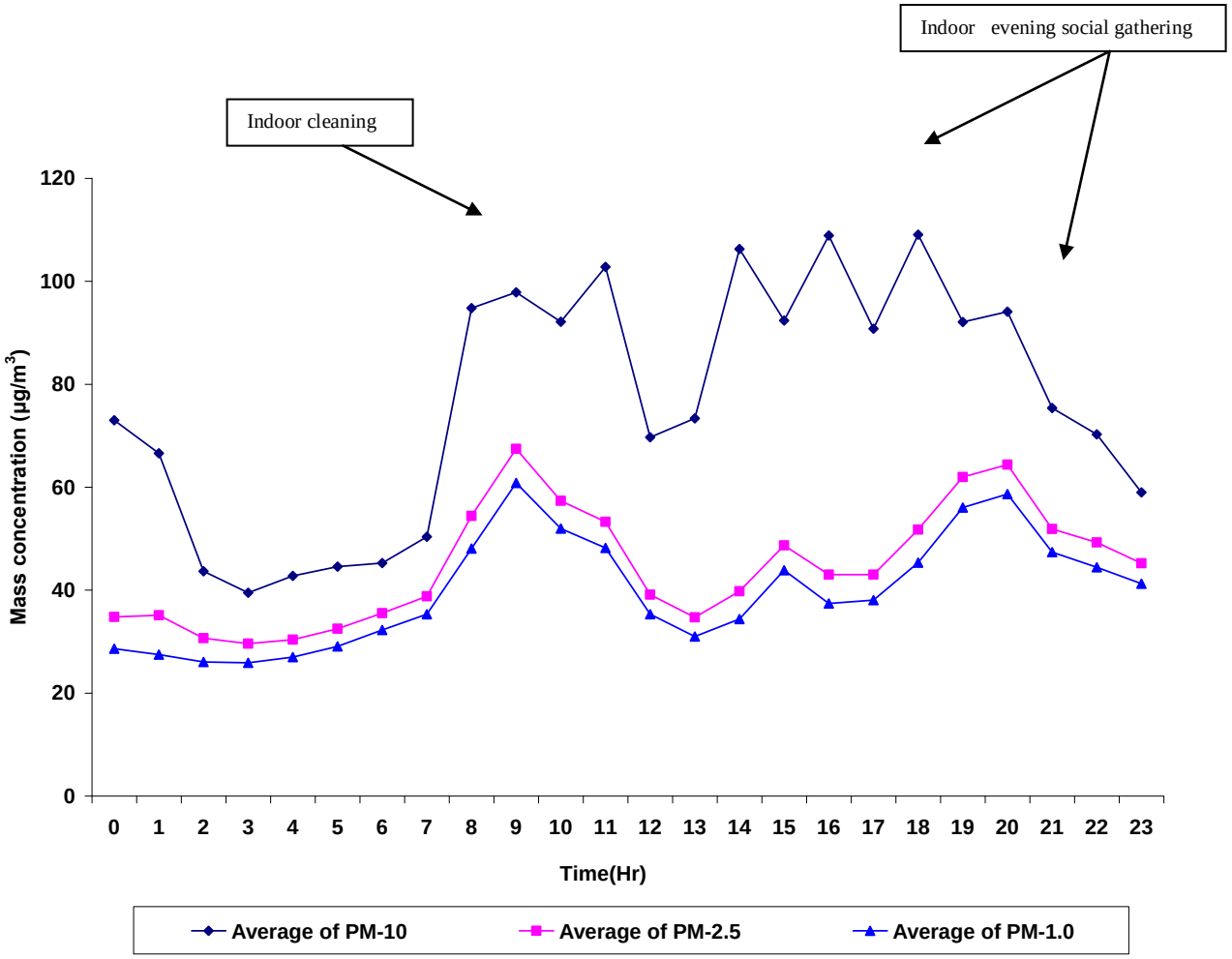

Figure 6. Representative hourly average mass concentration of PM_{10}, $PM_{2.5}$ and PM_1 in a non smoking living room living room at rural site in Pakistan on 12 – 01 – 06.

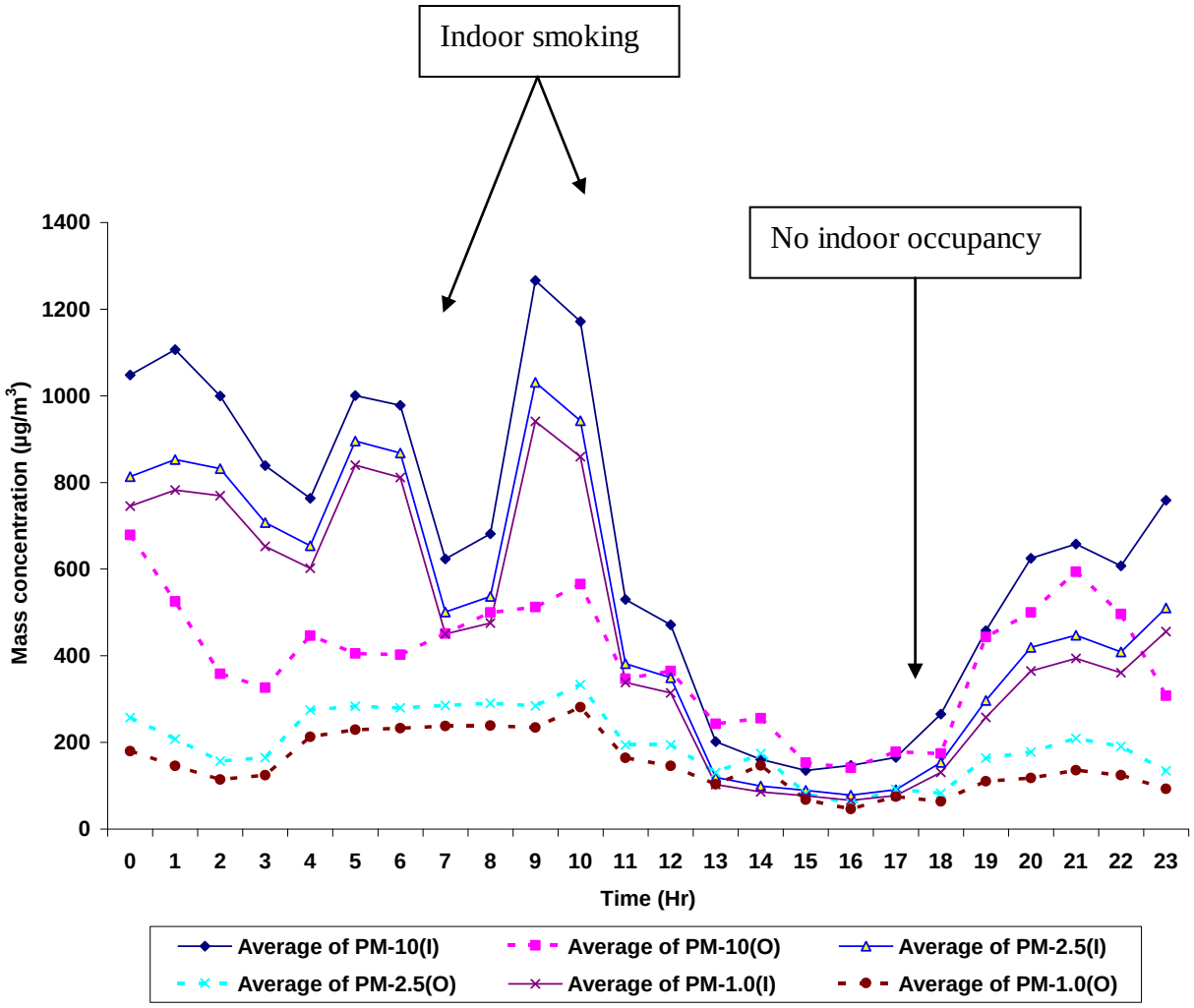

Figure 7. Representative hourly average of indoor and outdoor mass concentration of PM_{10}, $PM_{2.5}$ and PM_1 in a living room at an urban site (Lahore) in Pakistan on 04 -12 -05. I = Indoors; O = Outdoor