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Variation of Indoor Particulate Matter Concentrations and Association with Indoor/Outdoor Temperature: A Case Study in Rural Limpopo, South Africa

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Abstract: There is still a pressing concern regarding the causes of poor indoor air quality and the consequent effects on health, because people spend a considerable amount of time indoors. Information about seasonal variation and the determinants of particulate matter (PM) concentrations could guide the design and implementation of intervention strategies. This study was conducted in Giyani, Limpopo province, South Africa. The main aim was to assess indoor air quality. Indoor PM and temperature were monitored to describe seasonal and diurnal patterns of indoor PM₄ concentration and to estimate the association between PM concentrations and indoor as well as ambient conditions. Indoor PM₄ was monitored hourly in kitchens for the duration of spring (September), summer (February) and winter (July). Indoor temperatures were monitored hourly in kitchens, living rooms and bedrooms. Outdoor temperature and outdoor relative humidity were also monitored for the same period. Indoor temperatures showed a large range in the three sampled seasons, with the maximum values raising the largest cause for concern. Maximum indoor temperatures in summer exceeded the threshold of 35 °C, which has been shown to have adverse health effects. Occupants of the sampled households were exposed to indoor PM₄ concentrations that exceeded national and international guidelines. Hourly indoor temperature was statistically significantly correlated to PM₄ concentrations in the summer and spring (r = 0.22 and 0.24 respectively, p < 0.001 for both) and negatively correlated to outdoor relative humidity (r = -0.27, p < 0.001). Diurnal PM₄ variations showed pronounced patterns with morning and evening peaks. PM₄ was consistently higher throughout the day in summer compared to spring and winter. Community-based intervention strategies should consider these seasonal differences in PM4 exposure and tailor awareness messages for exposure prevention accordingly.

Keywords: air quality; diurnal variation; indoor; temperature; rural; South Africa

1. Introduction

The burning of wood, charcoal, animal dung and crop wastes to fulfill primary energy requirements is common around the world, with approximately 2.4 billion people relying on these solid fuels for their daily domestic requirements [1]. South Africa is no exception, with the national census data showing that at least 4,502,709 people use wood, coal or animal dung (also referred to as biomass fuel) as fuel for domestic cooking and space heating. A substantial proportion (26%) of these

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citizens reside in Limpopo province where wood is the most commonly used fuel for both cooking and space heating. Table 1 describes the proportions of people in Limpopo Province classified according to the sources of energy or fuel used for cooking and space heating as recorded in 2011 during the last national census.

|--|

	Number of Households (N = 14,450,161)						
	Electricity	Gas	Paraffin	Wood	Coal	Animal dung	Solar
Cooking Space heating	708,924.46 637,815.69	21,958.04 11,765.17	58,473.23 26,084.63	616,311.92 541,947.42	6381.00 12,594.70	1882.99 2420.50	1366.63 1940.52

During combustion, solid fuels release high concentrations of particulate matter (PM), carbon monoxide (CO), hydrocarbons, oxygenated organics, free radicals and chlorinated organics, which affect indoor environmental air quality [2–4]. The PM component of smoke released from burning biofuels is classified according to its aerodynamic diameter into size fractions such as PM_{10} (particles $\leq 10~\mu m$), PM_4 (particles $\leq 4~\mu m$), $PM_{2.5}$ (particles ≤ 2.5) and PM_1 , which consists of ultrafine particles which are $\leq 1~\mu m$ in size [5].

The concentration of PM in homes is dependent on various factors including building structure, types of human activities, the opening and closing of doors and meteorological factors such as temperature, wind, rainfall and humidity [6]. Differences in temperature indoors and outdoors influence natural ventilation through the movement of air. A higher rate of air exchange dilutes the concentration of PM generated indoors [6]. Changes in temperature also affect PM by influencing the change of chemical reaction rates and atmospheric mixing heights that affect the vertical dispersion of pollutants and modifying local wind and flow patterns that control the transportation of pollutants [7].

Studies conducted in rural areas have shown that the use of wood and/or coal for cooking or heating can be a significant source of particulate matter inside homes [8,9]. PM_{10} in homes that use biomass fuel was found to be 10–70 times above ambient concentrations observed in some of the world's most polluted cities [10]. Therefore, poor indoor air quality due to the combustion of biomass or fossil fuels can pose significant health risks due to exposure to increased levels of pollutants such as particulate matter [11].

Indoor conditions represent an important micro-environment because of the amount of time people spend in this space. Therefore, the main aim of this study was to assess indoor air quality in a rural setting among houses in Giyani, Limpopo Province, South Africa. Indoor PM₄ concentrations (known as the thoracic fraction of inhaled particles), indoor and outdoor temperature were monitored during spring, summer and winter. The objectives were to (1) assess the indoor temperatures and PM that household occupants are exposed to; (2) describe seasonal and diurnal patterns of indoor PM₄ concentration; and (3) estimate the association between PM concentrations and indoor as well as ambient temperature and relative humidity conditions.

The case study approach was applied to test the methods for a larger study that will include seasonal and spatial variability in four villages over two years. The findings reported here will be useful for the larger study, as well as several other studies presently underway across the country, for example, in Umlazi (KwaZulu-Natal Province) and Agincourt (Mpumalanga Province).

2. Materials and Methods

2.1. Study Site

The study was conducted in Giyani, a predominately rural town in the Mopani District, Limpopo Province of South Africa. The district has five sub-districts known as local municipalities, namely Greater Tzaneen, Greater Giyani, Greater Letaba, Maruleng and Ba-Phalaborwa. This study is one component of a larger project focused on determining environmental health in four villages in Limpopo.

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Air pollution, specifically indoor air pollution, is a major environment-related health threat due to the associated health risks of exposure. The four villages in the study are Tomu, Ka-Siyandani, Ka-Maswanganyi and Dzingidzingi. Two houses from each village were included in this air quality study (Figure 1). The households in Ka-Maswanganyi were sampled over three seasons (spring 2016, summer and winter 2017) while the households in the remaining three villages were sampled over two seasons (summer and winter 2017).

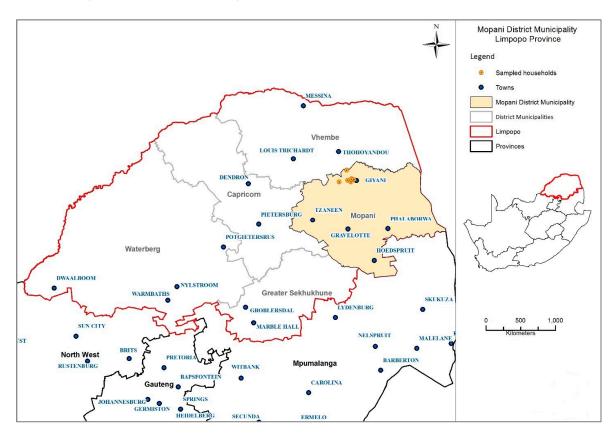


Figure 1. Map showing the location of Mopani District in Limpopo Province, South Africa (map produced in-house using ArcGIS[®] software (version 10.3) created by ESRI (Environmental Systems Resource Institute, Redlands, CA, USA)).

2.2. Household Questionnaire

In July 2017, a household survey was conducted as part of the larger project. A total of 408 structured questionnaires were administered to consenting participants representing the households. The responses relevant to the seven households used in this study were extracted from the larger dataset to be reported upon. Residents of the eighth household (located in Dzingidzingi) were unavailable at the time of the survey. The questions and responses are provided in detail in Appendix A. These pertain to type of fuel used for cooking and heating purposes and socio-demographic information.

2.3. Physical Indoor Air Quality Monitoring

Indoor air quality was monitored in spring, summer and winter in Giyani. Photometric light scattering instruments were used to continuously measure the concentration of respirable particulates within the indoor environment of individual households [12,13]. The DustTrak II Model 8530 has a laser with a wavelength of 780 nm, a mass concentration range of 0.001–100 mg·m $^{-3}$, and a particle size range of 0.1 to $\pm 10~\mu m$. Each instrument was flow calibrated, using a cyclone calibration jar and wet cell Sensidyne Gilian Gilibrator II, to a flow rate of 1.7 L·min $^{-1}$. This specific flow rate was selected as the instruments were fitted with a 10 mm Nylon Dorr-Oliver Cyclone, which provides a 50% cut size

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of 4 μ m (TSI Inc., Shoreview, MN, USA, 2013, 2014). Prior to the sampling, the instruments underwent zero calibration, flow calibration, filter changes and the cyclones were cleaned. These instruments were placed in the kitchen area of each house (they were not placed in the outside burning hut/kitchen where the household did also have such a facility).

The PM data was combined into a single data set and corrected by a calibration factor of 0.78. The calibration factor was obtained by comparing the continuous measurements to 24-hour gravimetric measurements sampled during the sampling period in the same households. This was done because previous studies that used the DustTrak [14–22] for photometric aerosol monitoring have indicated a significant overestimation of the particulate concentrations when compared to a reference gravimetric method. These studies were all conducted in various settings and compared to different reference methods. It is therefore critical to estimate a calibration factor for each monitor within the specific sampling environment.

2.4. Indoor and Outdoor Temperature Measurements

Thermochron iButton loggers were used to measure indoor and outdoor temperature. One logger was placed in the kitchen, bedroom and living room of each house as a representation of the temperature in the indoor environment. Loggers recorded measurements for the duration of spring (15 days in September), summer (23 days in February) and winter (29 days in July). A logging interval of 10 min was selected for each 24-hour period. Loggers were placed at a height of 1.6 m to ensure uniformity of sampling between the houses. Data was collected at the end of each sampling campaign and formatted into a single dataset.

2.5. Household Characteristics

Households in Giyani on average comprise five people. Household characteristics are presented in Appendix A. The most commonly used fuel for cooking was wood (five households) as opposed to electricity (two households). Most homes used wood fires for cooking in the morning (six households) and evening (four households) daily. The majority of households used wood or coal stoves (four households) and fireplaces (three households) to heat their homes during cold weather in winter. Only one household used an electric heater for space heating. None of the households reported using gas or paraffin for cooking or space heating. The eight sampled homes were constructed using bricks and all households were naturally ventilated through windows and doors.

2.6. Data Analysis

Statistical analysis were performed using R (version 3.4.2), a software environment for statistical computing and graphics (R Core Team, 2013). These analyses included descriptive statistics as well as exploring seasonal and diurnal variations. Pearson's correlation coefficient (r) was used to assess the relationship between PM₄ concentration and indoor and outdoor temperature and outdoor relative humidity.

3. Results

3.1. Seasonal Variation of Indoor Air Quality and Indoor and Outdoor Meteorological Variables

Hourly averages were calculated for ambient and indoor temperatures, ambient humidity and indoor PM₄ concentrations (Table 2). During the sampling campaigns, ambient temperatures ranged from 8.3 °C to 44.9 °C, compared to the indoor temperatures, which had warmer minimum and cooler maximum temperatures (ranging from 15.1 °C to 39.9 °C). Temperatures above 35 °C have been associated with heat-related illnesses and in extreme events, deaths [23,24]. Even though the maximum indoor temperatures were cooler than the ambient maximum temperature, they still exceeded this threshold value. External humidity ranged between 7.1% and 100%, with the highest humidity values recorded in the hottest months (February and March). The highest PM₄ concentration, 628 μ g·m⁻³,

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was measured in winter. Mean precipitation and wind speed were highest in summer (6.3 mm and 12.6 km/h, respectively).

Figure 2 shows hourly PM_4 concentrations in the eight kitchens sampled during the three seasons. The highest mean values were measured during spring (September). The highest variability was observed during winter (July), with a minimum of $1.9~\mu g \cdot m^{-3}$ and a maximum value of $628~\mu g \cdot m^{-3}$, measured at 6 p.m. Concentrations display seasonal variation with a significantly higher PM_4 measured in winter (July) and spring (September), compared to summer. This could be attributed to the fact that the majority of households reported in the questionnaire responses that they use biomass fuels for space heating during cold seasons. Another possible explanation for these levels of PM could be regional biomass burning that occurs during winter and spring [25]. Results obtained in previous studies found that PM in homes increased during cold seasons, when heating is common and ventilation is reduced, because doors and windows are typically closed, thus trapping PM inside [26,27].

The South African National Department of Environmental Affairs has legislated the National Ambient Air Quality Standards (NAAQS). These are 24-hour average recommendations, $65 \, \mu g \cdot m^{-3}$ for $PM_{2.5}$ and $75 \, \mu g \cdot m^{-3}$ for PM_{10} [28,29]. These standards are specific to outdoor air quality, there are no published standards or guidelines for indoor air quality in households in South Africa. The World Health Organization (WHO) also has ambient (outdoor) guidelines for 24-hour mean $PM_{2.5}$ and PM_{10} concentrations, which are $25 \, \mu g \cdot m^{-3}$ and $50 \, \mu g \cdot m^{-3}$, respectively [30]. Figures 3–5 show daily average indoor PM_4 concentrations measured in kitchens in all households compared to the $PM_{2.5}$ and PM_{10} concentration standards and guidelines set by NAAQS and the WHO, respectively. Levels of indoor PM_4 on all days in spring and winter exceeded the levels set by the WHO and NAAQS. However, in summer, none of the national daily average recommendations of PM_{10} and $PM_{2.5}$ by NAAQS were exceeded. The WHO international recommendations are exceeded on only a few days.

Variable	September (Spring 2016)		February (Summer 2017)		July (Winter 2017)	
variable	Mean (SD)	Range	Mean (SD)	Range	Mean (SD)	Range
Kitchen temperature (°C)	25.1 (3.2)	19.5–32.5	28.0 (3.3)	22.1–39.9	22.3 (2.1)	16.3–29.5
Bedroom temperature (°C)	25.0 (3.2)	19.5-32.3	28.5 (3.5)	22.4-38.4	21.5 (2.3)	15.6-27.8
Living room temperature (°C)	25.2 (3.4)	19.4-33.4	28.2 (3.4)	22.3-38.5	21.2 (2.1)	15.1-27.2
Outdoor temperature (°C)	24.9 (6.1)	15.9-44.9	26.7 (5.6)	19.0-43.0	19.3 (5.8)	8.3-35.3
Outdoor humidity (%)	38.3 (15.1)	7.5-87.9	73.4 (19.9)	16.4-100.0	54.9 (19.7)	7.1-93.1
PM4 ($\mu g \cdot m^{-3}$)	67.2 (42.0)	6.5-240.6	19.9 (25.7)	0.0 - 197.1	50.4 (51.9)	1.9-628.0
* Precipitation (mm)	0.3 (1.1)	0.0-7.9	6.3 (11.8)	0.0 - 28.0	0.4 (1.9)	0-10.9
* Wind speed (km/h)	11.8 (3.7)	6-21.0	12.6 (4.9)	6.0-24.0	10.5 (3.5)	3-19.0

Table 2. Descriptive statistics for indoor air quality and indoor and outdoor meteorological variables.

^{*} Obtained from Tzaneen weather station.

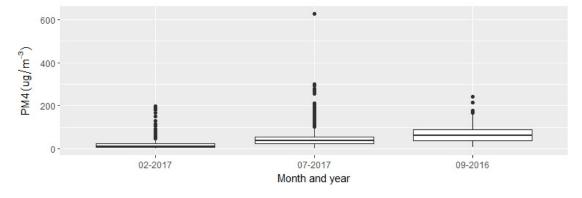


Figure 2. Box plot of average concentrations of PM₄ from all houses sampled in Giyani during 2016 and 2017 (**line**: median; **interquartile range box**: middle 50% of the data; **whiskers extending from either end of the box**: ranges for the bottom and top 25% of the data values; **solid black dots**: outliers).

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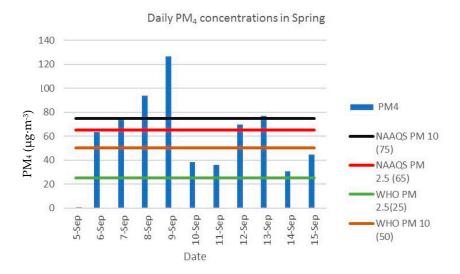


Figure 3. Indoor kitchen PM_4 concentrations measured in spring compared to NAAQS (National Ambient Air Quality Standards) standards and WHO guidelines.

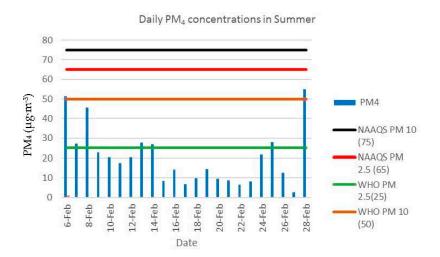


Figure 4. Indoor kitchen PM_4 concentrations measured in summer compared to NAAQS standards and WHO guidelines.

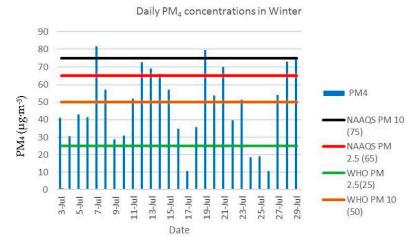


Figure 5. Indoor kitchen PM₄ concentrations measured in winter compared to NAAQS standards and WHO guidelines.

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Correlation Analysis

Table 3 presents Pearson's correlation coefficients and statistical significance between the outdoor temperature, indoor temperature, outdoor relative humidity and indoor PM₄ concentration. Statistically significant, although modest, positive correlations were found between indoor temperature and indoor PM₄ in spring and summer (r = 0.24, r = 0.22 respectively, p < 0.001 for both). Therefore, as indoor temperatures increase in spring and summer, so do indoor PM₄ concentrations. A significant negative correlation was observed between PM₄ and humidity in spring, although the strength of association was small (r = -0.27, p < 0.001). Therefore, as the outdoor relative humidity increases, the indoor PM₄ concentrations decrease. Precipitation and wind speed were not included because they were not monitored as part of this case study campaign; measurements were obtained from the closest weather station.

Table 3. Pearson's correlation coefficients showing the correlation between PM₄, temperature and relative humidity, stratified by season.

Variable	Spring	Summer	Winter
variable		Indoor PM ₄	
Indoor temperature	0.24 ***	0.22 ***	0.03
Outdoor temperature	0.11	0.04	-0.06
Outdoor relative humidity	-0.27 ***	-0.08	0.01

Note: Significance levels are indicated with * where * p < 0.05, ** p < 0.01, *** p < 0.001; indoor PM₄ and temperature were measured in kitchens.

3.2. Time Series Analysis

Diurnal Variation

Hourly PM_4 concentrations from the eight sampled households were averaged for spring, summer and winter. A pronounced bimodal diurnal pattern in PM concentrations was observed in all three seasons (Figure 6). PM_4 concentration peaked between 5 a.m. and 7 a.m. and again between 6 p.m. and 8 p.m. during all the seasons. However, PM_4 was consistently lower throughout the day in summer compared to spring and winter. The lowest concentrations of PM_4 were measured during the early hours of the morning before 5 a.m., between noon and 4 p.m. and during late evening for all seasons. Six of the eight households reported lighting fires using wood or coal in the morning and four in the evening. The results show that the morning and evening peaks correspond to these times of the day during which combustion and space heating occur.

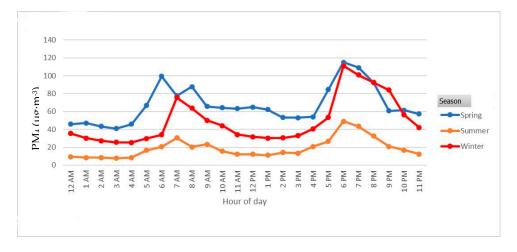


Figure 6. Diurnal time series of average hourly PM₄ concentrations by time of day and season.

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4. Discussion

Maximum indoor temperatures recorded in summer in the bedrooms, living room and kitchen exceeded 35 $^{\circ}$ C. A study conducted by Nitschke et al. [23] in South Australia found that for every 1 $^{\circ}$ C increment in maximum temperature above 35 $^{\circ}$ C, ambulance callouts increased by 0.7% and mortality increased by 0.9%. Household occupants were therefore exposed to adverse health effects due to elevated indoor temperatures.

The PM measured in this study, namely PM₄ is classified as the respirable fraction. This is the mass fraction of inhaled particles that penetrates to the unciliated airways [31]. It is particularly important because it can penetrate beyond the terminal bronchioles into the gas-exchange region of the lungs [32]. The inhalation of PM in indoor air causes harmful health effects because it deposits in the nasal, pharyngeal and laryngeal regions of the respiratory system, causing respiratory disease [33]. Therefore, it is important to understand the indoor and outdoor factors that affect indoor particle concentrations at sites that are representative of population exposure.

The range of the effects that PM has on human health is broad, but it predominately affects the respiratory and cardiovascular systems. The WHO guidelines and NAAQS standards for 24-hour mean PM_{10} are $50~\mu g \cdot m^{-3}$ and $75~\mu g \cdot m^{-3}$, respectively [28,30]. The average daily indoor PM_4 concentrations measured in the households in winter and spring exceeded the WHO guidelines and NAAQS standards for 24-hour mean PM_{10} , which are $25~\mu g \cdot m^{-3}$ and $65~\mu g \cdot m^{-3}$, respectively. Other studies conducted in low income countries also revealed that indoor PM concentrations were substantially higher than WHO guidelines [3,34,35]. This study confirms that household occupants are exposed to poor indoor air quality and that their health and comfort is likely being negatively impacted. An exposure response study in rural Kenya found that study participants exposed to high concentrations of PM experienced a higher frequency of acute respiratory infections [36].

We found a positive correlation between indoor PM₄ and indoor temperature in spring and summer. Several studies [37,38] found positive relationships between PM and temperature. This occurs because warm weather induces the formation of secondary fine particles, therefore increased temperatures result in increased PM [39,40]. PM was negatively correlated with relative humidity in spring. A similar study by Fromme et al. [41] also found a significant negative correlation between humidity and PM when assessing indoor air quality. This could have been caused by low relative humidity increasing particle deposition of fine particles [42,43] and high relative humidity decreasing particle deposition [44].

Most households cooked in kitchen structures that were not part of the main house, except for one household. These outdoor structures were however generally less than 100 m from the main house. Therefore, PM from combustion could have contributed to the PM measured inside the households. Furthermore, the majority of sampled households used wood and coal for space heating inside the homes. The kitchen inside the house is mostly used as a common area where space heating occurs; the burning of wood and coal exposes household occupants to fine particles in the air [45]. The diurnal pattern of PM concentrations coincides with the daily cycle of human activities associated with cooking and space heating. Diurnal variations in PM₄ concentrations measured in indoor kitchens showed morning and evening peaks, these peaks were during hours when heating using coal and wood usually occurs. The timing of these peaks is of particular concern because they occur during times when most people, including vulnerable groups, are inside the home. The number of people in a home and activities in the home such as smoking and dust resuspension by cleaning also contribute to higher PM levels [26].

Since this was a case study, these results require validation in a large sample and in households located in other rural locations. One study limitation was that the only instrument available to measure PM measured PM $_4$. Despite the national standards and international air quality guidelines being for PM $_{2.5}$ and PM $_{10}$, we used these to evaluate our PM $_4$ concentrations as an indication of the possible presence of health risks due to PM exposure.

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Our results suggested a strong seasonal variability in PM, with variability being highest in winter. Exposure to extreme PM values was thus highest in winter. A study by Wheeler et al. [46] found that both indoor and outdoor 24-hour-averaged PM concentrations were significantly higher in winter than in summer, by as much as 50%. Research has attributed this seasonal variation to several factors, including the longer residence time of particulates in the atmosphere during winter due to low winds and low mixing height [47,48] and wood and coal burning for heating [49,50]. Poor ventilation in winter could also have contributed to the increased PM concentrations observed in the study, similar observations were reported by Nasir et al. [51]. Elevated concentrations of PM observed in spring during our study could be attributed to regional biomass burning. Biomass burning emissions contribute significantly to the aerosol burden in a region [52]. Studies suggest that the maximum biomass burning activity in South Africa occurs between June and September [25,53].

5. Conclusions

Indoor temperatures showed a large range in the three sampled seasons, with the maximum values raising the largest cause for concern. Maximum indoor temperatures in summer exceeded the threshold of 35 °C, which has been shown to have adverse health effects. Indoor temperature was found to have a statistically significant positive effect on indoor PM concentrations. Indoor household concentrations of PM4 exceeded the standards and guidelines set by the NAAQS and the WHO, respectively. The implications of these findings are that increased heating in homes, either from lighting fires for cooking, or for heating, reduces indoor air quality by increasing the concentration of PM indoors. Also, the high indoor PM4 concentrations observed in this study could have been associated with higher background concentrations. Information about seasonal variation and the determinants of PM concentration could guide the design, timing and implementation of intervention strategies. Effective household indoor air quality monitoring programs may need to include interventions in seasons other than winter, for example, spring, when indoor air quality may also be poor. As a result, this could reduce exposure to PM and limit the associated health risks to people living in households using biomass or fossil fuels.

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Author Contributions: C.Y.W. conceived the air quality study and prepared the study protocol. B.L. implemented the fieldwork. T.K. and B.L. performed the data analyses. All authors contributed to the writing of the manuscript.

Conflicts of Interest: The authors declare there are no conflicts of interest.

Declarations: Permission to install the air quality monitoring equipment and temperature and RH loggers in the households was obtained from Limpopo Provincial Department of Health and the Mopani District Municipality. Research ethics clearance was granted from the South African Medical Research Council Research Ethics Committee (EC005-3/2014).

Appendix

N = 7	
Question	Count of responses
How many people including you make up this main household?	
4–6	5
6–8	2
>8	-

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How many children under the age of 5 are part of this household?	
0	4
1–3	3
>3	-
How many people over the age of 60 are part of this household?	
0	3
1–3	4
>3	-
During very cold weather, do you use any of the following systems to heat	
your home?	
Wood or coal stove	4
Fireplace	3
Gas	-
Electric heater	1
Paraffin heater	0
How often do you use the wood/coal stove?	
About everyday	4
2–3 times a week	
2–3 times a month	
Seldom	
Never	
How often do you use the fireplace?	
About everyday	3
2–3 times a week	· ·
2–3 times a month	
Seldom	
Never	
How often do you use the electric heater	
About everyday	
2–3 times a week	
2–3 times a month	1
Seldom	1
Never	
What fuel do you mainly use for cooking?	2
Electricity	2
Paraffin	
Gas	-
Wood	5
Coal	
Crop waste/cow dung	
Does the family sit in the same room around the stove or cooking fire—when cooking is taking place?	
Yes	
No	7
	,
Please specify when fires are made?	4
Morning	6
Midday	A
Evening	4
Where do you do your daily cooking?	
kitchen (part of the house)	1
kitchen (separated from the house)	6

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Do you have any dampness in the dwelling?		
No	7	
Yes	0	
Do you have any leaks in the roof or leaking water pipes in or around		
the dwelling?		
No	7	
Yes	0	

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