

International Journal of Environmental Research and Public Health

Article

Occupational Exposure to Fine Particulate Matter (PM₄ and PM_{2.5}) during Hand-Made Cookware Operation: Personal, Indoor and Outdoor Levels

Busisiwe Shezi ^{1,2,*}, Angela Mathee ^{1,2,3}, Nokulunga Cele ⁴, Sipho Ndabandaba ⁴ and Renee A. Street ^{4,5}

- ¹ Environment and Health Research Unit, South African Medical Research Council, Johannesburg 2094, South Africa; Angie.Mathee@mrc.ac.za
- ² Department of Environmental Health, Faculty of Health Sciences, University of Johannesburg, Johannesburg 2094, South Africa
- ³ Department of Environmental Health, Nelson Mandela University, Port Elizabeth 6019, South Africa
 ⁴ Environment and Health Research Unit, South African Medical Research Council, Durban 4001, South Africa;
- nokulungacele6@gmail.com (N.C.); ndabandabasp@gmail.com (S.N.); Renee.Street@mrc.ac.za (R.A.S.)
 ⁵ University of KwaZulu-Natal, Discipline of Occupational and Environmental Health, School of Nursing and Public Health, Durban 4001, South Africa
- * Correspondence: busisiwe.shezi@mrc.ac.za

Received: 8 September 2020; Accepted: 5 October 2020; Published: 16 October 2020



Abstract: (1) Exposure of informal artisanal cookware makers to fine particles has not yet been characterized. The aim of this study was to characterize occupational exposure to fine particulate matter (PM_4 and $PM_{2.5}$) levels and fine particulate matter ($PM_{2.5}$) elemental components; (2) Artisanal cookware makers were recruited from five cookware making sites. Exposure to fine particulate matter was measured for 17 male participants. SidePak personal aerosol monitors (AM520) were used to measure personal exposure to PM_4 , while a DustTrak monitor and an E-sampler were used to assess indoor and outdoor $PM_{2.5}$ levels, respectively. A questionnaire was administered to capture information on demographic characteristics. The chemical characterization of indoor and outdoor $PM_{2.5}$ filter mass was conducted using Wavelength Dispersive X-ray Fluorescence. Time series record of 15-min averages for indoor and outdoor $PM_{2.5}$ levels were assessed; (3) The median (range) was 124 µg/m³ (23–100,000), 64 µg/m³ (1–6097) and 12 µg/m³ (4–1178), respectively, for personal $PM_{4,}$ indoor and outdoor $PM_{2.5}$ filter mass and (4). The information generated during this study may assist in extending occupational health and safety strategies to artisanal cookware makers and developing targeted prevention initiatives.

Keywords: hand-made cookware operation; aluminum; personal exposure; microenvironment assessment; cottage industry; informal sector; fine particles

1. Introduction

Epidemiological studies have consistently found significant positive associations between airborne particulate matter (PM) and a variety of adverse health outcomes, including respiratory and cardiovascular illnesses [1–3] and adverse pregnancy outcomes [4–7]. According to the World Health Organization, PM affects more people than any other pollutant [8]. In addition, long term health effects of exposure to PM outweigh short term health effects with regards to public health significance [9]. PM is defined by the United States Environmental Protection Agency as "a mixture of solid and liquid droplets suspended in the air such as organic chemicals, soot, metals, soils or dust



particles and biological material" [10]. PM may contain a mixture of several pollutants distributed at various sizes including PM_{0.1} (aerodynamic diameter less than 0.1 micrometers), PM_{2.5} (aerodynamic diameter less than 2.5 micrometers) or PM₄ (aerodynamic diameter less than 4 micrometers) [11].

In occupational settings, exposure to PM may arise from, for example, agricultural activities [12], boiler making [13], metal mining [14–17], and informal, home-based industrial operations [18]. Informal, home-based industries include spray painting [19], welding [20–22], electrical appliance repairs [23] and hand-made cookware operations [24,25]. Recently, concerns have been raised about the harmful contents of locally available hand-made cookware [26,27]. For example, studies conducted in Cameroon [26] and South Africa [27] have reported the use of toxic metals during the hand-made cookware operation, including lead and cadmium. Similar findings have been reported in West African countries, i.e., Nigeria [24,25]. However, there is a dearth of information about exposure to toxic substances among cookware makers during the hand-made cookware making process. Artisanal cookware makers use scrap metal including used car and motorbike engine parts, waste aluminum and computer components to make cookware [24,25]. It is well known that PM and its elemental components are virtually always present in particle-generating processes, especially combustion processes [28]. For example, during hand-made cookware operations, PM and elemental components are produced through the combustion process to cast liquid aluminum into cookware [24,26]. In addition, cookware makers are exposed to the latter during the sand mold production, solidification monitoring and removing and trimming the casting [25,29].

Occupational health and safety is essential for the well-being of both formal (regulated, registered and protected by the labor legislation) and informal (not recognized, registered, regulated and/or protected under labor legislation and social protection) workers [30]. However, the use of administrative controls (i.e., job rotation), engineering controls and personal protective equipment is not always practiced in informal settings [31]. Further, little attention has been given to the effects of occupational exposures, which differ from general environmental exposures in both particle type (e.g., composition), as well as exposure frequency (e.g., environmental exposures are relatively constant while occupational exposures are more variable), duration, intensity or levels (e.g., occupational exposures are generally higher than environmental exposures). These differences in exposure composition, duration, frequency and population may have implications on how occupational exposures impact health.

There are limited occupational health studies where personal, indoor and outdoor PM have been characterized and compared, especially among informal workers, and none to our knowledge in artisanal cookware makers. In order to get a better understanding of the health risks posed by PM among artisanal cookware makers, we measured occupational exposure to fine particulate matter (PM_4 and $PM_{2.5}$) during hand-made cookware operation and assessed the chemical characterization of indoor and outdoor $PM_{2.5}$ filter mass. The relationship between personal, indoor and outdoor PM levels was also examined.

2. Materials and Methods

2.1. Study Design, Setting and Population

This study was undertaken during the months of June and July 2019. The target population was artisanal cookware makers situated in the province of Limpopo (Giyani) and Kwa-Zulu Natal (Durban), South Africa. Giyani is a city in the north-eastern region of the Limpopo province of South Africa and Durban is situated along the east coast of the country. Signed informed consent was obtained from all participants. The study was approved by the University of KwaZulu-Natal Biomedical Research Ethics Committee (reference number: BE410/18).

2.2. Exposure Assessment

Seventeen artisanal cookware makers were assessed for personal exposure to PM₄. Five hand-made cookware operation sites were monitored for PM_{2.5} (Figure 1 shows one of the hand-made cookware

operation sites); however, outdoor and/or indoor measurements were incomplete in one site (there was no electricity to connect the E-sampler and unwillingness to participate for the entire duration of indoor sampling period led to incomplete measurements). A questionnaire was administered to capture demographic characteristics such as age, marital status, tobacco smoking and education. The questionnaires were captured face-to-face, by field workers. Study data were collected and managed using REDCap electronic data capture tools.



Figure 1. One of the hand-made cookware operation sites with artisans preparing sand mold.

2.3. Personal Air Monitoring

A portable, real time photometric aerosol monitor (SidePakTM TSI model AM520, TSI Inc., Shoreview, MN, USA) was used to measure PM₄ levels among 17 adult males making artisanal cookware. The monitor was carried by the cookware makers for a period of 3 h at a flow rate of 1.7 L/min. The breathing zone of participants was assessed using a 10-mm Nylon Dorr-Oliver Cyclone inlet (TSI Inc., Shoreview, MN, USA) which differentiates between the respirable fraction and other portions of PM. The cyclone was attached to the worker's clothing near his head. The logging interval was set at 1 min. The SidePak monitors were calibrated prior to the data collection phase using the guidelines set by the manufacturer.

2.4. Indoor Air Monitoring

A DustTrak (TSI Inc., Shoreview, MN, USA) photometric light scattering monitor which had been calibrated one year before the sampling period was used to measure indoor $PM_{2.5}$. Both real time and mass sampling (using mixed cellulose ester membrane filters, 37 mm (SKC Ltd, Dorset, UK)) were used for sampling indoor $PM_{2.5}$ (for a period of 8 h). The sampler flow rate was set at 3 L/min using a 1 min logging interval. A suitable location (at least 0.5 m from the wall and away from the door or gap between the wall and the roof) was identified for placing the DustTrak monitor.

2.5. Outdoor Air Monitoring

An E-Sampler (Met One Instruments, Grants Pass Oregon) was used to assess outdoor $PM_{2.5}$ for a period of 8 h. The E-sampler is designed to measure the amount of scattered light and uses a conversion factor (indicated by K) to convert light scattering to mass. Therefore, both real time (1-min logging interval) and mass sampling were conducted. Mixed cellulose ester membrane filters, 47 mm (SKC Ltd, Dorset, UK) were used for sampling outdoor $PM_{2.5}$. The sampler flow rate was set at 2 L/min. The E-sampler calculated the K constant during operation and self-adjusted itself. The E-sampler has internal temperature and relative humidity sensors to autocorrect for high atmospheric water content, which can influence the measurements.

2.6. Gravimetric Analysis

Gravimetric analysis was used to weigh filters prior to sampling using an XP26 DeltaRange Microbalance (Mettler-Toledo AG, Greifensee, Switzerland) accurate to 1 microgram. The 37- and 47-mm filters were conditioned for 24 h in the laboratory with controlled indoor climate of 22 °C \pm 2 °C and relative humidity (not captured). After sample collection, the filters were returned to the petri dishes, conditioned, re-weighed and stored at 4 °C until they could be chemically analyzed. The PM_{2.5} levels were calculated using the following equation:

$$PM_{2.5} \ \mu g/m^3 = ([(Wf-Wi)-blank \ filter \ mass] \times 10^{-6})/Vt$$
(1)

where Wf is the filter mass after sampling and Wi is the filter mass before sampling in grams (g). The blank filter is the change in mass of the blank filter calculated by weighing the blanks before and after sampling and is expressed in grams (g). Vt is the total volume of sampled air during the 8-h period. Total volume was calculated using the sampled flow rate, ambient temperature and pressure during sampling. Each site had around (915 L (0.9 m³): Esampler), and (1395 L (1.4 m³): DustTrak) sampled air over the 8-h period. The resulting concentration was calculated in $\mu g/m^3$.

2.7. PM_{2.5} Chemical Characterization

For additional information on elemental composition of the indoor and outdoor PM_{2.5} filter mass, the filters were subsequently analyzed for 49 elements at the University of North West, South Africa, using wavelength-dispersive x-ray fluorescence (WD-XRF) technique. The following elements were analyzed Ag, Al, As, Au, Ba, Bi, Br, Ca, Ce, Cd, CI, Co, Cr, Cs, Cu, Fe, Ga, Ge, Hg, I, In, K, Li, Mg, Mn, Mo, Na, Nb, Ni, P, Pb, Pd, Pt, Rb, S, Sb, Sc, Se, Si, Sn, Sr, Te, Ti, Tl, V, W, Y, Zn and Zr.

2.8. Quality Assurance and Control

The SidePak and DustTrak monitors were factory calibrated according to the ISO 12103-1, A1 Arizona test dust. Prior to data collection, the latter instruments were zero calibrated by attaching the zero-filter, as recommended by the manufacturer. The batteries were charged on a daily basis to maintain battery capability during sampling and the impactor plates were checked and cleaned on a daily basis. The output for personal, indoor and outdoor measurements was given in milligram per cubic meter (mg/m³) but converted to microgram per cubic meter (μ g/m³) prior to data analysis. Data were downloaded from the instruments after every sampling period using the TSI Trackpro (DustTrak and SidePak) and Comet (E-sampler). A standardized log sheet was used to record sampling start and stop times. Laboratory and field blank filters (indoor (n = 4), outdoor (n = 4)) were used to adjust the weight difference observed due to changes in indoor climate of the weighing room and field handling of samples, respectively. The field blanks (indoor (n = 2), outdoor (n = 2) were taken to the hand-made cookware operation sites and were handled the same way as the other filters (i.e., the field blank filters were loaded into the sampler for five minutes (no air was drawn through the blank filters). The limit of detection (LOD) was estimated using the method by Vaughan et al., [32] using the mean of standard deviation of the mass change of blank filters multiplied by three. Standardized field sampling flow rates for the E-sampler and DustTrak were used. The LOD divided by the square root of 2 was used in all calculations [33]. All indoor and outdoor values were above the limit of detection. Invalid PM levels were identified by inspection of plots; a small number of cases when the monitor appeared to be performing inaccurately were noted. Zero and negative values were excluded from the dataset. The indoor and outdoor $PM_{2.5}$ photometric measurements were adjusted by using the calibration factor to approximate the actual PM_{2.5} mass levels.

3. Statistical Analysis

All data were cleaned and checked for quality on Microsoft Excel and exported to Stata IC version 14 (StataCorp, College Station, TX, USA) for further analysis. Statistical analysis was restricted to those

observations that had concurrent levels of personal, indoor and outdoor air pollution measurements available. Descriptive statistics, scatterplots, and histograms were used to characterize distributions of PM levels. Quartiles, means (standard deviations) and medians (range) for PM were used. Real time data were collected every 1 min, but 15-min averages were used for analysis.

The 17 workers who were assessed for 3-h personal sampling were categorized into three groups: (i) 1st PM₄ sampling sessions; workers assessed for personal sampling in the morning, (ii) 2nd PM₄ sampling sessions; workers assessed for personal sampling just before midday, and (iii) 3rd PM₄ sampling sessions; workers assessed for personal sampling in the afternoon. Concurrent 3-h measurements of indoor and outdoor PM_{2.5} measurements were matched with the 3-h PM₄ personal measurements. Four workers were excluded from this analysis because of incomplete/no PM_{2.5} measurements to match with PM₄ levels. The relationship between 3-h personal, indoor and outdoor measurements was assessed using Spearman's rank order correlation.

The relationship between indoor and outdoor PM_{2.5} levels was examined using time series indoor-outdoor plots and by calculating Spearman's rank order correlation using the 15-min averages of PM.

4. Results

4.1. Demographic and Site Characteristics

All 17 cookware makers were male. The mean (SD) for participant age was 34 years (11). The majority (59%) were single and had attended or completed high school (65%). Wood was used in three of the five sites and in two sites artisans used coal for melting aluminum. All sites had a gap between the wall and the roof and were built with corrugated metal sheeting.

Two sites were situated in low socio-economic urban areas and three in rural areas. Four of the five sites were on residential plots. Descriptions of the sites sampled and the number of workers who participated in PM sampling are summarized in Table 1.

Site No.	Total No. of Workers	1st PM ₄ Sampling Session	2nd PM ₄ Sampling Session	3rd PM ₄ Sampling Session	Indoor Sampling	Outdoor Sampling	Volume of the Site (m ³)
1	5	2	1	-	Yes	Yes	42.5
2	2	2	-	-	Yes	Yes	34.9
3	5	2	-	-	Yes	Yes	77.8
4	13	2	2	2	Yes	Yes	127.0
* 5	6	2	2	-	Yes (incomplete)	No	71.7

Table 1. Description of the sites and workers sampled.

* There was no electricity to connect the E-sampler and unwillingness to participate for the entire indoor 8-h sampling period led to incomplete measurements.

4.2. Real Time Personal, Indoor and Outdoor PM Measurements

PM levels in our study were high (Table 2). The real time personal PM₄, indoor and outdoor PM_{2.5} levels were negatively skewed. The levels ranged from 23 to 100,000 μ g/m³; 1 to 6097 μ g/m³ and 1 to 1178 μ g/m³, respectively, for personal PM₄, indoor and outdoor PM_{2.5} levels. When comparing personal PM₄ and indoor and outdoor PM_{2.5} levels, personal exposure levels were found to be the highest, mean (SD) was 492 μ g/m³ (3546), with a median of 124 μ g/m³, followed by workplace indoors, mean (SD), 98 μ g/m³ (262), with a median of 64 μ g/m³. The mean (SD) and median for workplace outdoors was 20 μ g/m³ (45) and the median was 13 μ g/m³ (Table 2).

Levels	Personal PM ₄ (µg/m ³)	Indoor PM _{2.5} (µg/m ³)	Outdoor PM _{2.5} (µg/m ³)	Outdoor Temperature (°C)	Outdoor Humidity (%)
Min	23	1	3.6	16	6
10th percentile	74	31	7	18	15
25th percentile	90	44	9	23	22
50th percentile	124	64	13	26	27
75th percentile	182	99	19	28	36
Max	100 000	6097	1178	38	55
Mean (SD)	492 (3546)	98 (262)	20 (45)	26 (5)	29 (11)

Table 2. Particulate matter (PM) measurements and meteorological data.

Abbreviations: SD (standard deviation); µg/m³ (micrograms per cubic meter).

4.3. Indoor and Outdoor PM_{2.5} Concentration Measurements

The mass measurements for indoor and outdoor PM_{2.5} levels ranged from 6 to 371 μ g/m³ and 8.8 to 51 μ g/m³, respectively. The mean (SD) was 105 μ g/m³ (137), and 19 μ g/m³ (16); and the median was 61 μ g/m³ and 14 μ g/m³, respectively, for indoor and outdoor measurements.

4.4. Meteorological Data

Outdoor temperature was mainly in the range of 16 to 38 °C, with a mean (SD) of 26 °C (5). Relative humidity ranged from 6% to 55% with a mean (SD) of 29% (11) (Table 2).

4.5. Personal, Indoor and Outdoor Relationships (3-h Measurements)

Personal–indoor and personal–outdoor Spearman correlations (*r*) of 3-h concurrent measurements were poor for both the 1st and 2nd PM sampling session. Personal–indoor correlations and personal–outdoor correlations were (r = 0.05, p > 0.05), (r = -0.18; p > 0.05), and (r = -0.08, p > 0.05), (r = -0.37, p > 0.05), respectively, for workers sampled in the 1st and 2nd PM sampling session.

4.6. Indoor and Outdoor Relationships (8-h Measurements)

Correlations between indoor–outdoor $PM_{2.5}$ samples, indoor $PM_{2.5}$ –outdoor meteorological factors and outdoor $PM_{2.5}$ –outdoor meteorological factors are shown in Table 3. Indoor–outdoor $PM_{2.5}$ correlations were moderate for site 2 (r = 0.67, p < 0.0001). For site 1, 2 and 3 the indoor–outdoor $PM_{2.5}$ correlations were (r = -0.32, p < 0.05), (r = 0.28, p > 0.05) and (r = 0.43, p < 0.05), respectively.

Site No.	Indoor/Outdoor PM _{2.5} Levels	Indoor PM _{2.5} /Temperature	Outdoor PM _{2.5} /Temperature	Indoor PM _{2.5} /Humidity	Outdoor PM _{2.5} /Humidity
1	-0.32	0.08	-0.15	-0.07	0.37
2	0.67	-0.73	-0.73	0.80	0.84
3	0.28	-0.39	-0.85	0.33	0.88
4	0.43	-0.53	-0.48	0.51	0.50

Table 3. The Spearman's rank order correlation for 15-min averages of indoor and outdoor PM_{2.5} levels, outdoor temperature and outdoor humidity.

Bolded values indicate significance at 95% confidence interval (95% CI).

For all sites, outdoor PM_{2.5} levels were negatively correlated with outdoor temperature. This correlation ranged from weak (r = -0.15) to high (r = -0.85). Correlation between indoor PM_{2.5} levels and outdoor temperature was also negatively correlated in three sites. The correlation was moderate (r = -0.39, p > 0.05) to high (r = -0.73, p < 0.05). All sites had a positive correlation between microenvironmental levels and humidity except an insignificant weak negative correlation between indoor PM_{2.5} levels and humidity observed in site 1 (r = -0.07, p > 0.05).

4.7. Time Series Record of 15-min Average Indoor and Outdoor PM_{2.5} Real Time Measurements

The relationship between indoor and outdoor $PM_{2.5}$ real time levels is shown in Figure 2. In all sites indoor $PM_{2.5}$ levels remained higher than outdoor levels with little evidence of strong outdoor sources. There were frequent large indoor peaks reflecting significant indoor sources. Across all sites average indoor 15-min levels were variable than the outdoor levels.



Figure 2. A comparison of time series record of 15-min average indoor and outdoor $PM_{2.5}$ real-time measurements (**a**) Site 1: five cookware makers occupied this site, and it was situated in rural areas and wood was used in furnaces; (**b**) Site 2: two cookware makers occupied this site, and it was situated in rural areas and wood was used for furnace; (**c**) Site 3: thirteen workers occupied this site, and it was situated in the urban area and coal was used for furnace; (**d**) Site 4: 6 workers occupied this site, and it was situated in urban areas and wood was used for furnace.

4.8. PM_{2.5} Elemental Components

Out of 49 indoor and outdoor analyzed elements, thirteen (Ag, Au, Bi, Co, Ga, Ge, Hg, Li, Mo, Nb, Se, Sn, and Tl) had zero values and were excluded in the results. The results for 38 elements are shown in Table 4. The highest levels for many of the elemental components of $PM_{2.5}$ were found in the outdoor filter mass (Table 4). The elemental components for both indoor and outdoor $PM_{2.5}$ were much lower than the occupational exposure limit recommended by the South African regulations for Hazardous Chemical Substances and Lead regulations under the Occupational Health and Safety (OHS) Act (Act 85 of 1993).

Sample Name	Indoor (µg/m ³)	Outdoor (µg/m ³)	*8-h TWA OEL-RL (μg/m ³)
Al	0.2	1.9	10500
As	1.0	2.5	100
Ba	0.1	0.2	500
Br	0.0	0.2	100
Ca	1.4	1.4	-
Ce	0.4	0.9	-
Cd	0.1	0.0	50
CI	1.9	3.2	-
Cr	0.4	0.6	50
Cs	0.3	0.5	
Cu	4.0	7.1	1000
Fe	0.9	1.9	-
Ι	0.2	0.4	-
In	0.9	0.7	-
Κ	0.3	2.9	-
Mg	0.1	1.3	5100
Mn	0.1	0.6	5000
Na	7.7	7.4	-
Ni	0.4	0.8	50
Р	0.0	0.1	100
Pb	2.8	6.6	150
Pd	71.8	162.2	-
Pt	12.2	28.7	5000
Rb	0.2	0.2	-
S	0.2	0.1	-
Sb	1.9	4.8	-
Sc	0.1	0.2	-
Si	0.8	7.4	100
Sr	0.2	0.0	-
Te	8.2	18.3	100
Ti	0.0	0.1	-
V	0.0	0.1	-
W	0.0	0.1	5000
Y	0.0	0.2	1000
Zn	5.3	9.7	
Zr	0.2	0.1	5000

Table 4. Metal levels of indoor and outdoor $PM_{2.5}$ filter mass.

* TWA OEL-RL: Time Weighted Average Occupational Exposure Limit-Recommended Limit.

Some of the PM elements are not reported in the regulation for Hazardous Chemical Substances under the OHS Act (Act 85 of 1993).

5. Discussion

In this study, we described personal (PM_4), indoor and outdoor ($PM_{2.5}$) levels and $PM_{2.5}$ elemental components among artisanal cookware makers. The relationship between personal, indoor and outdoor measurements was assessed. Our data are consistent with the general pattern observed in most PM monitoring studies [34–37]: outdoor levels are lower than indoor levels, and both indoor and outdoor levels are lower than personal levels. Elemental components in outdoor $PM_{2.5}$ filter mass are higher than indoors. These findings strengthen our understanding of the worker–exposure relationship during hand-made cookware operations, especially with the substantial differences observed between personal, indoor and outdoor measurements.

Particulate matter is associated with respiratory effects and chronic obstructive pulmonary diseases [2,3,38] and has been reported to result in increased hospital admissions, outpatient visits and emergency admissions [39,40]. Other studies have reported an inverse association between

particulate matter and adverse pregnancy outcomes such as birth defects, low birth weight and preterm delivery [41,42]. Overall, the levels of fine particles found in our study were high. The air quality guidelines for PM_{2.5} suggested by the WHO expert group are $10 \,\mu\text{g/m}^3$ as an annual mean and $25 \,\mu\text{g/m}^3$ as a 24-h mean [43]. In this study we obtained mean levels of 98 and $20 \,\mu\text{g/m}^3$ for indoor and outdoor measurements (8-h mean) of PM_{2.5}, respectively. We compared our personal measurements with the existing occupational exposure limits in South Africa. According to the regulations for Hazardous Chemical Substances, under the OHS Act (Act 85 of 1993), the personal exposure limit for respiratory dust is 5000 $\mu\text{g/m}^3$ (average 8-h respiratory dust). In our study, the 3-h time weighed average concentrations ranged from 42 to 300 $\mu\text{g/m}^3$; therefore, they were much lower than the occupational exposure limit. However, in site 5 we observed very high concentrations of PM₄ (ranging between 5010 and 100,000 $\mu\text{g/m}^3$), this might have been because coal was used for furnace in this site.

Exposure of artisanal cookware makers during cookware making is undocumented in the literature. Some studies conducted previously have shown that microenvironment assessment does not always reflect personal exposure [34–36]. Similarly, concurrent samples undertaken in this study revealed that the averaged personal exposure levels (3-h mean) at breathing zones of workers are much greater (490 μ g/m³) than those of the microenvironment levels. Though the comparisons were based on two different fractions of PM (PM₄ and PM_{2.5}) and despite the undocumented exposures among artisanal cookware makers, other studies have reported similar findings [34,36]. The high personal PM₄ levels observed in this study might be related to frequent mobility of the workers within their working areas. Furthermore, various activities of the workers such as design and production of the mold for the cookware, pouring of melted aluminum into the mold cavity, solidification monitoring, removing and trimming the casting [25,29] could significantly increase the PM levels.

In all the sites, indoor $PM_{2.5}$ levels remained higher than outdoor levels with little evidence of strong outdoor sources. However, we also observed considerable differences between the sites, for example, frequent large indoor peaks were observed in some of the sites. This could be explained by the differences in activity levels, the number of workers per site and the size of the working site (which ranged from 35 to 127 m³). In addition, natural ventilation in all the sites was in the form of a gap between the wall and the roof, allowing particles from outside to readily penetrate into the workplace. Therefore, another possible explanation for higher indoor levels may be the infiltration of outdoor pollutants indoors, which may be influenced by wind speed and wind direction [44].

In our study, the cookware making sites were situated on residential plots, and therefore exposed groups may include the entire family, neighbors and the cookware makers. Therefore, the measurements of indoor and outdoor $PM_{2.5}$ levels provided an indication of how the workers are exposed to $PM_{2.5}$ and the potential impacts this might have to the families living in these sites.

A weak correlation was observed between personal–indoor and personal–outdoor levels. This could be explained by the variations in time spent indoors and outdoors by the workers, differences in ventilation and distance between the cookware making area and furnace across the sites. Similar findings have been observed from other studies comparing indoor, outdoor and personal measurements in occupational settings [45,46].

Epidemiological studies have reported that particle-bound metals are associated with mitochondrial damage [47] and induction of oxidative stress [48], which results in an increase in cardiovascular mortality and morbidity [49]. These elements may originate from various industrial or urban sources. In our study area, the mean outdoor metal levels were higher than indoors. This may be as a result of the outdoor furnace, where various types of metal containing products are used, i.e., e-waste (from computer parts), vehicle and motor bike engine parts. Even so, when comparing our study findings with the occupational exposure limit reported under the Hazardous Chemical Substances Regulation (1995) and Lead Regulations (2001) of the OHS Act (Act 85 of 1993), our results were much lower.

One of the study limitations was that a convenience sample of artisanal cookware makers was drawn, which limits our ability to generalize our findings. In our study, different sampling equipment

was used for personal, indoor and outdoor PM levels. A good agreement between the real-time measurements and the particle mass measurements was shown by the overall distribution of PM_{2.5} mass levels, which was comparable to the distribution of real-time PM_{2.5} levels. Unlike levels of PM_{2.5} and PM_{10} , concurrent levels of PM_4 and $PM_{2.5}$ have rarely been investigated in workplaces. By taking parallel measurements of PM₄ and PM_{2.5} we were able to compare levels of the two size fractions; however, $PM_{2.5}$ fraction is a fraction of PM_4 , so the quantities cannot be compared equivalently. Because of the small sample size, we could not assess the association between PM and exposure variables such as smoking. However, in our study the number of participants who were active smokers was limited. Therefore, the impact of tobacco smoke on personal, indoor and outdoor levels is likely to be limited. Light scattering photometers have been reported to result in overestimation of the PM concentrations when compared to a gravimetric method. In our study, we used light scattering photometers and gravimetric analysis. To obtain accurate gravimetric analysis of PM, temperature stability, extreme care exercise in sample handling and moisture control is required [50]. However, relative humidity was not captured by the laboratory for our study. Therefore, it is not clear whether the relative humidity was within the recommended limit or not during our study. The performance of PM monitors varies because of the differences in technology, therefore, using different monitors may limit the comparison of the same fraction of PM. In our study we used different instruments for measuring personal, indoor and outdoor PM.

6. Conclusions

This to our knowledge is the first study to assess PM during hand-made cookware operations. The levels measured at the breathing zone of the workers were higher than the levels from indoors and outdoors. There were frequent large indoor peaks reflecting significant indoor sources. The chemical characterization of indoor and outdoor PM filter mass provided metal levels and the mean outdoor elemental levels were relatively higher than mean indoor elemental levels. According to our results and those of many others, levels measured at microenvironment level are a poor predictor of personal (cross-sectional short-term) workday exposure (from all sources). Therefore, identification of occupational exposure during hand-made cookware operation allows for more inclusive occupational health and safety strategies.

Author Contributions: Conceptualization, A.M. and R.A.S. methodology, B.S. formal analysis, B.S. investigation, B.S., N.C; and S.N. resources, A.M and R.A.S. data curation, B.S., N.C. and S.N. writing—original draft preparation, B.S. writing—review and editing, B.S., A.M. and N.C. S.N. and R.A.S. supervision, B.S. and R.A.S. project administration, B.S., A.M., N.C., S.N. and R.A.S. funding acquisition, A.M. and R.A.S. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Research Foundation, Grant number 116068.

Acknowledgments: We would like to thank all the artisanal cookware makers who participated in this study. This work was supported by the South African Medical Research Council.

Conflicts of Interest: The authors declare no conflict of interest

References

- 1. Korten, I.C.S.; Ramsey, K.; Latzin, P. Air pollution during pregnancy and lung development in the child. *Paediatr. Respir. Rev.* **2017**, *21*, 38–46. [CrossRef]
- 2. Jafta, N.; Jeena, P.M.; Barregard, L.; Naidoo, R.N. Childhood tuberculosis and exposure to indoor air pollution: A systematic review and meta-analysis. *Int. J. Tuberc. Lung Dis.* **2015**, *19*, 596–602. [CrossRef]
- 3. Laumbach, R.J.; Kipen, H.M. Respiratory health effects of air pollution: Update on biomass smoke and traffic pollution. *J. Allergy Clin. Immunol.* **2012**, *129*, 3–11. [CrossRef]
- 4. Mulenga, D. Maternal exposure to household air pollution and associated adverse birth outcomes in Ndola and Masaiti, Zambia. *Ec. Pulmonol. Respir. Med.* **2018**, *7*, 82–97.

- Jedrychowski, W.A.; Majewska, R.; Spengler, J.D.; Camann, D.; Roen, E.L.; Perera, F.P. Prenatal exposure to fine particles and polycyclic aromatic hydrocarbons and birth outcomes: A two-pollutant approach. *Int. Arch. Occup. Environ. Heal.* 2017, 90, 255–264. [CrossRef] [PubMed]
- Khan, N.; Nurs, C.Z.B.; Islam, M.M.; Islam, A.Z.; Rahman, M. Household air pollution from cooking and risk of adverse health and birth outcomes in Bangladesh: A nationwide population-based study. *Environ. Heal.* 2017, *16*, 57. [CrossRef] [PubMed]
- 7. Naidoo, S.; London, L.; Burdorf, A.; Naidoo, R.; Kromhout, H. Spontaneous miscarriages and infant deaths among female farmers in rural South Africa. *Occup. Environ. Med.* **2011**, *68*, A51. [CrossRef]
- 8. WHO. Ambient (Outdoor) Air Quality and Health 2018. Available online: https://www.who.int/news-room/fact-sheets/detail/ambient-(outdoor)-air-quality-and-health (accessed on 7 September 2020).
- 9. WHO. Health Aspects of Air Pollution: Results from the WHO Project "Systematic Review of Health Aspects of Air Pollution in Europe"; WHO: Copenhagen, Denmark, 2004.
- 10. USEPA. Particulate Matter (PM) Pollution 2012. Available online: https://www.epa.gov/pm-pollution. (accessed on 7 September 2020).
- 11. Kelly, F.J.; Fussell, J.C. Size, source and chemical composition as determinants of toxicity attributable to ambient particulate matter. *Atmospheric Environ.* **2012**, *60*, 504–526. [CrossRef]
- Naidoo, S.; London, L.; Burdorf, A.; Naidoo, R.N.; Kromhout, H. Agricultural Activities, Pesticide Use and Occupational Hazards among Women Working in Small Scale Farming in Northern KwaZulu-Natal, South Africa. *Int. J. Occup. Environ. Heal.* 2008, 14, 218–224. [CrossRef]
- 13. Kile, M.L.; Fang, S.C.; Baccarelli, A.A.; Tarantini, L.; Cavallari, J.M.; Christiani, D.C. A panel study of occupational exposure to fine particulate matter and changes in DNA methylation over a single workday and years worked in boilermaker welders. *Environ. Heal.* **2013**, *12*, 47. [CrossRef]
- 14. Naidoo, R.N. Mining: South Africa's legacy and burden in the context of occupational respiratory diseases. *Glob. Heal. Action* **2013**, *6*, 20512. [CrossRef]
- Naidoo, R.N.; Robins, T.G.; Seixas, N.; Lalloo, U.G.; Becklake, M. Respirable Coal Dust Exposure and Respiratory Symptoms in South-African Coal Miners: A Comparison of Current and Ex-Miners. J. Occup. Environ. Med. 2006, 48, 581–590. [CrossRef]
- 16. Van Staden, J.; Foos, R.; Volckens, J.; Ramachandran, G. Variability of aerosol mass and number concentrations during taconite mining operations. *J. Occup. Environ. Hyg.* **2019**, *17*, 1–14. [CrossRef]
- Huynh, T.B.; Ramachandran, G.; Quick, H.; Hwang, J.; Raynor, P.C.; Alexander, B.H.; Mandel, J.H. Ambient Fine Aerosol Concentrations in Multiple Metrics in Taconite Mining Operations. *Ann. Work. Expo. Heal.* 2018, 63, 77–90. [CrossRef] [PubMed]
- Teare, J.; Kootbodien, T.; Naicker, N.; Mathee, A. The Extent, Nature and Environmental Health Implications of Cottage Industries in Johannesburg, South Africa. *Int. J. Environ. Res. Public Heal.* 2015, 12, 1894–1901. [CrossRef] [PubMed]
- 19. Hansen, K.T.; Vaa, M. *Reconsidering Informality: Perspectives from Urban Africa;* Nordic Africa Institute: Uppsala, Sweden, 2004.
- 20. Clausen, J.; Rastogi, S. Heavy metal pollution among autoworkers. *I. Lead. Occup. Environ. Med.* **1977**, *34*, 208–215. [CrossRef]
- 21. Clausen, J.; Rastogi, S.C. Heavy metal pollution among autoworkers. II. Cadmium, chromium, copper, manganese, and nickel. *Occup. Environ. Med.* **1977**, *34*, 216–220. [CrossRef]
- Lehnert, M.; Pesch, B.; Lotz, A.; Pelzer, J.; Kendzia, B.; Gawrych, K.; Heinze, E.; Van Gelder, R.; Punkenburg, E.; Weiss, T.; et al. Exposure to Inhalable, Respirable, and Ultrafine Particles in Welding Fume. *Ann. Occup. Hyg.* 2012, *56*, 557–567. [CrossRef]
- 23. Foulke, E. *Guidance for the Identification and Control of Safety and Health Hazards in Metal Scrap Recycling;* Occupational Safety and Health Administration, US Department of Labour OSHA: Washington, WA, USA, 2008; 3348-05.
- 24. Osborn, E.L. Casting aluminium cooking pots: Labour, migration and artisan production in West Africa's informal sector, 1945–2005. *Afr. Identit.* **2009**, *7*, 373–386. [CrossRef]
- 25. Guma, T.N.; Uche, L.O. Sand Mould Design for Casting an Aluminium Pot-A Basic Procedure of Supplementing Artisanal Practices. *Eur. J. Eng. Res. Sci.* **2019**, *4*, 24–30. [CrossRef]
- 26. Weidenhamer, J.D.; Kobunski, P.A.; Kuepouo, G.; Corbin, R.W.; Gottesfeld, P. Lead exposure from aluminum cookware in Cameroon. *Sci. Total. Environ.* **2014**, *496*, 339–347. [CrossRef]

- 27. Street, R.A.; Mathee, A.; Tanda, S.; Hauzenberger, C.; Naidoo, S.; Goessler, W. Recycling of scrap metal into artisanal cookware in the informal sector: A public health threat from multi metal exposure in South Africa. *Sci. Total. Environ.* **2020**, 699, 134324. [CrossRef]
- 28. Colbeck, I.; Nasir, Z.A.; Nasir, Z.A. *Indoor Air Pollution*; Springer Science and Business Media LLC: Berlin/Heidelberg, Germany, 2010; pp. 41–72.
- 29. Guma, T.; Uche, O.L. A typification of foundry practices for correct artisanal sand casting of aluminum pots. *Int. J. Eng. Appl. Sci. Technol.* **2019**, *4*, 169–178. [CrossRef]
- ILO. Informal Sector: ILO.; 2020. Available online: https://www.ilo.org/global/topics/dw4sd/themes/informaleconomy/lang--en/index.htm (accessed on 7 September 2020).
- 31. Amfo-Otu, R.; Agyemang, J.K. Occupational health hazards and safety practices among the informal sector auto mechanics. *Appl. Res. J.* 2017, *2*, 59–69.
- 32. Milligan, B.D.; Vaughan, N.P.; Ogden, T.L. Filter weighing reproducibility and the gravimetric detection limit. *Ann. Occup. Hyg.* **1989**, *33*, 331–337. [CrossRef]
- 33. Hornung, R.W.; Reed, L.D. Estimation of Average Concentration in the Presence of Nondetectable Values. *Appl. Occup. Environ. Hyg.* **1990**, *5*, 46–51. [CrossRef]
- 34. Lee, B.-K.; Smith, T.J.; Garshick, E.; Natkin, J.; Reaser, P.; Lane, K.; Lee, H.K. Exposure of trucking company workers to particulate matter during the winter. *Chemosphere* **2005**, *61*, 1677–1690. [CrossRef]
- Di Gilio, A.; Farella, G.; Marzocca, A.; Giua, R.; Assennato, G.; Tutino, M.; De Gennaro, G. Indoor/Outdoor Air Quality Assessment at School near the Steel Plant in Taranto (Italy). *Adv. Meteorol.* 2017, 2017, 1–7. [CrossRef]
- Kousa, A.; Oglesby, L.; Koistinen, K.; Kunzli, N.; Jantunen, M.J. Exposure chain of urban air PM2.5—Associations between ambient fixed site, residential outdoor, indoor, workplace and personal exposures in four European cities in the EXPOLIS-study. *Atmospheric Environ.* 2002, *36*, 3031–3039. [CrossRef]
- 37. He, C.; Morawska, L.; Hitchins, J.; Gilbert, D. Contribution from indoor sources to particle number and mass concentrations in residential houses. *Atmos. Environ.* **2004**, *38*, 3405–3415. [CrossRef]
- 38. Fisk, W.J.; A Eliseeva, E.; Mendell, M.J. Association of residential dampness and mold with respiratory tract infections and bronchitis: A meta-analysis. *Environ. Heal.* **2010**, *9*, 72. [CrossRef]
- Weichenthal, S.; Lavigne, E.; Evans, G.J.; Pollitt, K.J.G.; Burnett, R.T. Fine Particulate Matter and Emergency Room Visits for Respiratory Illness. Effect Modification by Oxidative Potential. *Am. J. Respir. Crit. Care Med.* 2016, 194, 577–586. [CrossRef]
- Weichenthal, S.; Villeneuve, P.J.; Burnett, R.T.; Van Donkelaar, A.; Martin, R.V.; Jones, R.R.; Dellavalle, C.T.; Sandler, D.P.; Ward, M.H.; Hoppin, J.A. Long-Term Exposure to Fine Particulate Matter: Association with Nonaccidental and Cardiovascular Mortality in the Agricultural Health Study Cohort. *Environ. Heal. Perspect.* 2014, 122, 609–615. [CrossRef]
- 41. Sun, X.; Luo, X.; Zhao, C.; Zhang, B.; Tao, J.; Yang, Z.; Ma, W.; Liu, T. The associations between birth weight and exposure to fine particulate matter (PM2.5) and its chemical constituents during pregnancy: A meta-analysis. *Environ. Pollut.* **2016**, *211*, 38–47. [CrossRef]
- Sun, X.; Luo, X.; Zhao, C.; Ng, R.W.C.; Lim, C.E.D.; Zhang, B.; Liu, T. The association between fine particulate matter exposure during pregnancy and preterm birth: A meta-analysis. *BMC Pregnancy Childbirth* 2015, 15, 300. [CrossRef]
- WHO. WHO Air Quality Guidelines Global Update 2005. In Proceedings of the Working Group Meeting, Bonn, Germany, 18–20 October 2005; WHO Regional Office for Europe: Copenhagen, Denmark, 2005.
- Wichmann, J.; Lind, T.; Nilsson, M.-M.; Bellander, T. PM2.5, soot and NO2 indoor–outdoor relationships at homes, pre-schools and schools in Stockholm, Sweden. *Atmospheric Environ.* 2010, 44, 4536–4544. [CrossRef]
- Van Drooge, B.L.; Rivas, I.; Querol, X.; Sunyer, J.; Grimalt, J.O. Organic Air Quality Markers of Indoor and Outdoor PM2.5 Aerosols in Primary Schools from Barcelona. *Int. J. Environ. Res. Public Heal.* 2020, 17, 3685. [CrossRef]
- 46. Chen, X.-C.; Chuang, H.-C.; Ward, T.J.; Tian, L.; Cao, J.-J.; Ho, S.S.-H.; Lau, N.-C.; Hsiao, T.-C.; Yim, S.H.; Ho, K.-F. Indoor, outdoor, and personal exposure to PM2.5 and their bioreactivity among healthy residents of Hong Kong. *Environ. Res.* **2020**, *188*, 109780. [CrossRef]

- Li, N.; Sioutas, C.; Cho, A.; Schmitz, D.; Misra, C.; Sempf, J.; Wang, M.; Oberley, T.; Froines, J.; Nel, A. Ultrafine particulate pollutants induce oxidative stress and mitochondrial damage. *Environ. Heal. Perspect.* 2003, *111*, 455–460. [CrossRef]
- 48. E Nel, A.; Diaz-Sanchez, D.; Li, N. The role of particulate pollutants in pulmonary inflammation and asthma: Evidence for the involvement of organic chemicals and oxidative stress. *Curr. Opin. Pulm. Med.* **2001**, 7, 20–26. [CrossRef]
- 49. Araujo, J.A.; Nel, A.E. Particulate matter and atherosclerosis: Role of particle size, composition and oxidative stress. *Part. Fibre Toxicol.* **2009**, *6*, 24. [CrossRef] [PubMed]
- 50. ASTM, editor ASTM Standard Practice for Controlling and Characterizing Errors in Weighing Collected Aerosols (ASTM D 6552)2000. Available online: http://www.astm.org/Standards/D6552.htm (accessed on 28 September 2020).

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



© 2020 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).