Particle emissions from household fires in South Africa

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Abstract

The effects of airborne particles on the atmosphere, climate and public health are among the central topics in current environmental research. South Africa is a developing country, therefore not all households have access to electricity. In municipalities in the province of the Eastern Cape, for example, electricity usage for cooking ranges from 95.5% of households to only 8.1%. The households which do not utilise electricity make use of other resources to meet their needs in terms of energy provision for lighting, cooking and heating, including charcoal and wood burning in braziers. This paper investigates the particle emissions from household fires prepared using two methods, with reference to potential human health effects. Particles were sampled onto quartz fibre filters, which were analysed by scanning electron microscopy and GCxGC-MS. Particulate matter collected near the traditional fire contained higher concentrations of adsorbed organic compounds, including polycyclic aromatic hydrocarbons, which are of concern due to their potential human health effects. Changes in combustion conditions had a marked effect on the organic and particulate emissions. An integrated approach to reduce potential exposure from household fires should be explored, including energy efficient housing design; the provision of cost effective, energy efficient combustion appliances; as well as appropriate community education campaigns.

Keywords: particle, denuder, household fires, quartz fibre filter, human health, polycyclic aromatic hydrocarbons, GCxGC-MS, SEM.

1 Introduction

The effects of airborne particles on the atmosphere, climate and public health are among the central topics in current environmental research. These particles



originate from numerous natural and anthropogenic sources, leading to a variation in chemical and physical composition of the particulate matter, which in turn leads to different human health and environmental effects.

South Africa is a developing country, therefore not all households have access to electricity. The provision of resources is being continuously improved, as is evident from comparison of the results of the census conducted in South Africa in 2001 to that of a community survey held in 2007. The overall percentage of households which use electricity for lighting increased from 69.7 to 80.0%; whilst electricity used for cooking purposes increased from 51.4 to 66.5%; and for heating from 49.0 to 58.8% [1]. It can be seen that more households use electricity for lighting than for cooking and even less utilise electricity for heating purposes.

The households which do not utilise electricity make use of other resources to meet their needs, including paraffin, charcoal and wood burning, which is of concern as the World Health Organisation (WHO) has identified indoor air pollution from solid fuel burning as one of the world's ten major causes of mortality and morbidity [2]. The WHO has estimated that exposure to biomass fuel smoke results in ~1.5 million premature deaths per year, with the most firmly established health effects of such exposure being acute lower respiratory tract infections in children and chronic obstructive pulmonary disease in adults [3].

This paper investigates the particle emissions from household fires, with reference to potential human health effects as well as possible mitigation measures which are relevant to developing countries. The effect of variations in combustion parameters on total suspended particulate emissions was investigated with respect to domestic fires. The particulate matter was characterised by means of scanning electron microscopy and comprehensive gas chromatography with mass spectrometric detection.

2 Electricity usage in South Africa

The magnitude of potential human health effects from exposure to domestic fire emissions generally show seasonal variations, due to the increased use of domestic fires for heating purposes during the winter months. Besides temporal fluctuations, geographical variations in potential exposure in South Africa are obvious when the provincial and municipal electricity usage data is studied. Fig. 1 shows the distribution of electricity usage across the nine provinces of the country, the locations of which are shown in Fig. 2. It is evident that provinces with more developed commercial centres, such as Gauteng and the Western Cape, have more consistent (with respect to usage for lighting, cooking as well as heating) and higher usages than that of more rural provinces, such as the Eastern Cape. In some provinces, such as Limpopo, electricity usage for cooking and heating is much lower than that for lighting purposes.

These geographical differences in terms of usage of electricity can be further seen in Fig. 3, which gives an indication of the range of electricity usage for lighting across the municipalities in each province and how these compare to the



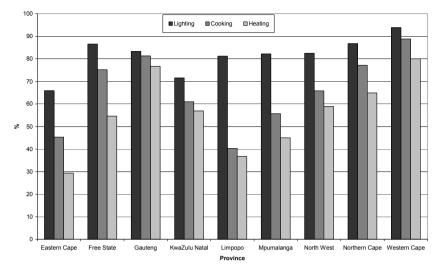


Figure 1: Percentage electricity usage for lighting, cooking and heating per province of South Africa (adapted from [1]).



Figure 2: Provincial map of South Africa.

national average usage. It can be seen that the majority of the municipalities in each province exceed the national average, with the exception of the Eastern Cape and KwaZulu Natal. However, in five out of the nine provinces, a greater percentage of municipalities have electricity usages for lighting which are below the average usage for that province. In these cases, the provincial average is elevated by relatively few municipalities which have high usages.

The disparity is even more pronounced when electricity usage for cooking purposes is considered (Fig. 4). Here the majority of municipalities in only four of the provinces (Free State, Gauteng, Northern Cape and Western Cape) had electricity usages higher than the national average. It is of concern that in five of

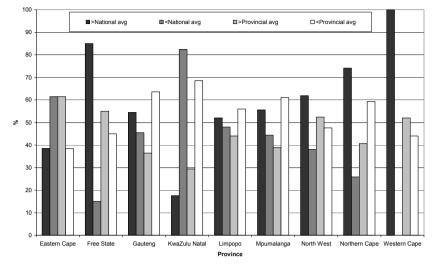
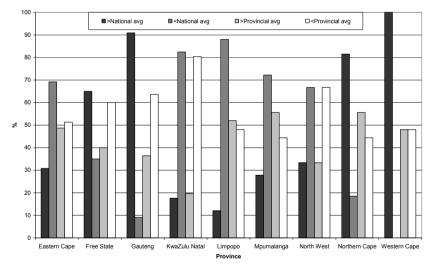
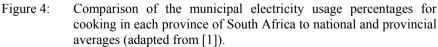


Figure 3: Comparison of the municipal electricity usage percentages for lighting in each province of South Africa to national and provincial averages (adapted from [1]).





the provinces, a greater proportion of municipalities had electricity usages for cooking below the average for that province.

The wide range of electricity usage across municipalities in the different provinces is shown in Fig. 5, which shows the municipality with the maximum

percentage of households using electricity for cooking purposes as well as the municipality with the minimum percentage usage for each province. In the Eastern Cape, for example, 95.5% of the households use electricity for cooking in the municipality with the maximum electricity usage, whilst only 8.1% of households use electricity for cooking in the municipality with the lowest electricity usage. The latter municipality has a population of 165 084 people, which represents ~0.3% of the population of South Africa. However, it can be argued that the potential exposure of these people to atmospheric emissions from household fires is still significant.

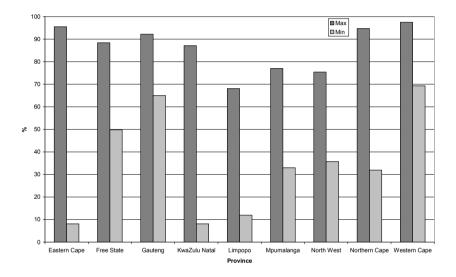


Figure 5: Comparison of the municipalities with the highest and lowest electricity usage percentages for cooking in each province of South Africa (adapted from [1]).

This data illustrates the need to understand the potential human health risks as a result of particulate emissions from household fires, due to the portion of the population which do not use electricity for lighting, cooking and heating purposes. Residential combustion of D-grade coal has been found to be the greatest source of particulate matter in a township in the Free State, for example, contributing 62.1% of PM_{2.5} and 42.6% of PM₁₀ [4].

3 Experimental method

3.1 Sampling

Fires were made in braziers, which are homemade batch combustion appliances commonly used in South Africa for both cooking and heating purposes. In each combustion test, the appliance was initially at room temperature. Fires were prepared using the following two methods.



3.1.1 Traditional method

Four pieces of rolled up newspaper were placed on a grid at the bottom of a brazier, followed by 250 g of wood shards and then 100 g of logs. 3 kg of commercial charcoal was placed above this and the fire was lit. Air samples were taken by means of a portable sampling pump ~60 cm above the brazier at a flow rate of 0.5 L.min⁻¹. Samples were simultaneously taken at a distance of 2.5 m from the brazier at a flow rate of 1 L.min⁻¹. Particulates were sampled 10 min after ignition of the fire for 30 min onto pre-cleaned quartz fibre filters (6 mm diameter), held between two empty glass tubes with a Teflon connection. Gas phase organic emissions were sampled for 10 min onto two polydimethylsiloxane (PDMS) multi-channel rubber traps [5] in series (primary and secondary traps, respectively) 40 min after ignition.

3.1.2 "Basa njengo Magogo" (BnM) method

This "upside down" method of lighting a fire is recommended for domestic use due to a reduction in air emissions, including total particulates [6]. The composition and size of these particles and the nature of organic emissions have not been investigated, however.

Due to the larger capacity of the brazier used here, fuel masses were doubled compared to method 2.1.1 but the ratio was kept the same. 5 kg of charcoal was placed in the brazier, followed by 4 pieces of newspaper, 800 g of logs and 200 g of wood shards. After ignition of the fire, 1 kg of charcoal was added. Gas and particle phase emissions were collected by a miniature denuder, consisting of two silicone rubber traps in series with a quartz fibre filter between them [7, 8], at 1 L.min⁻¹ for 10 min (Fig. 6).



Figure 6: Sampling of gas and particle phase emissions by means of a PDMS/quartz fibre filter/PDMS denuder above the brazier.

3.2 Analysis

3.2.1 Scanning Electron Microscopy (SEM) with Energy Dispersive X-Ray Spectroscopy (EDS)

The quartz fibre filters were sputter coated with gold prior to analysis by SEM (JSM 5800LV) and EDS (Thermo Scientific, operating at 20 kV).

3.2.2 TD-GCxGC-TOFMS

Comprehensive two-dimensional gas chromatography with time of flight mass spectrometric detection (GCxGC-TOFMS, LECO Pegasus 4D) was employed in the analysis of thermally desorbed (Gerstel) filter samples (particle associated organic emissions) and silicone rubber traps (gas phase organic emissions). Helium was used at a desorption flow rate of 50 mL.min⁻¹ and analytes were cryogenically focussed at -40°C. The primary column was a 20 m RTx 5SilMS (180 μ m id; 0.18 μ m film thickness) and the secondary column was a 1.5 m RTx 200 (180 μ m id; 0.2 μ m film thickness). The modulation period was 5 s.

4 Results and discussion

Typical morphologies of collected particles as determined by SEM are shown in Fig. 7, and were similar for both combustion methods. Both irregularly shaped porous char particles and spherical cenospheres were evident. The formation of condensation aerosols on the fibres of the filter were found for the BnM burn, which may indicate higher combustion temperatures compared to the traditional method. The nature of the fuel, the air/fuel ratio, as well as the temperature and duration of the combustion process all play a role in determining the morphologies of the particulate products [9].

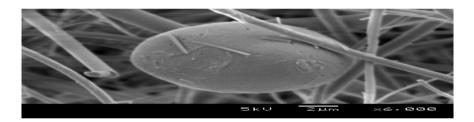


Figure 7: SEM images showing morphologies of particles collected onto quartz fibre filters (a) irregularly shaped porous char, (b) cenosphere with a smooth surface, and (c) condensation aerosols formed on the filter fibres during the BnM burn.

The EDS analyses revealed that the cenospheres and condensation aerosols only contained carbon and oxygen, which indicates that they may have been formed by the same vaporization-condensation processes during combustion. The char particles, on the other hand, displayed a different elemental profile, as they contained relatively less carbon and more oxygen, as well as inorganic elements arising from the fuel (Fe, Mg and K).

Due to the vast number of analytes determined by TD-GCxGC-MS, those for which the signal to noise ratio was <300:1, the library match was <80% and the peak area was <100 000 were discarded, as were PDMS breakdown products, which would have arisen from the PDMS trap material. The contribution of

Table 1:Particle phase organic compounds determined by TD-GCxGC-MS
analysis of filter samples taken at a traditional brazier fire and
2.5 m from it. The peak area for filter 2 was adjusted in order to
account for the larger sample volume.

| Filter # | Filter 1 | | Filter 2 | |
|---|-------------------------|-------------|-----------------------|-------------|
| Sampling position | At brazier | | 2.5 m from brazier | |
| Sampling flow rate | 0.5 L.min ⁻¹ | | 1 L.min ⁻¹ | |
| | % total peak area | Peak area | % total peak area | Peak area/2 |
| Alcohols | 12.1 | 94 380 177 | 10.8 | 35 338 490 |
| Acids | 2.3 | 17 988 790 | 6.3 | 20 810 642 |
| Aldehydes | 1.5 | 11 548 373 | 2.4 | 8 015 354 |
| Ketones | 7.9 | 61 986 660 | 3.6 | 11 867 201 |
| Esters | 16.1 | 125 806 222 | 12.2 | 40 228 271 |
| N-containing non- heterocyclic | 9.8 | 76 931 117 | 6.8 | 22 288 912 |
| Heterocyclic | 10.1 | 79 211 731 | 19.6 | 64 220 195 |
| Chlorinated & fluorinated compounds | 13.3 | 103 645 428 | 12.2 | 40 206 522 |
| PAHs | 1.0 | 7 730 632 | 0.2 | 701 441 |
| Derivatives of PAHs | 0.5 | 4 067 332 | 0.1 | 213 262 |
| Other | 25.4 | 198 548 399 | 25.8 | 84 600 635 |

different classes of chemical compounds to the adjusted total peak area was then determined.

It is evident upon comparison of peak areas, that the particulate matter collected near the traditional fire contained higher concentrations of adsorbed organic compounds (including polycyclic aromatic hydrocarbons (PAHs) and PAH derivatives, which are of concern due to their potential human health effects), as would be expected due to dilution effects further from the fire (Table 1). The PAHs which were detected were primarily phenanthrene, fluoranthene and pyrene. Organic acids were present at slightly higher concentrations further from the fire, based on peak areas, which may be products of atmospheric oxidation reactions. A higher proportion of heterocyclic compounds were found associated with particulates further from the fire (Fig. 8),



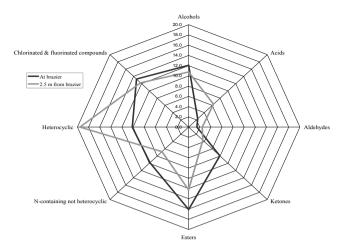


Figure 8: Variation in relative composition of selected particle phase organic compound emissions from a traditional brazier fire at the fire and 2.5 m away from it, based on the percentage contribution of different chemical classes to the total adjusted peak area.

which may have resulted from condensation and ring formation reactions of compounds emitted from the fire. Levoglucosan, which is a tracer for biomass burning, was found in the particulate samples.

Gas phase organic emissions are an important consideration from a human health perspective in addition to that associated with the particle phase, as the fate of the two phases in the human lung may be different. The contribution of vapour phase PAH concentrations to the total PAHs emitted from residential coal stoves in China, has been reported to be significant [10]. Gas phase compounds may also become adsorbed onto emitted particulate matter in the combustion plume as it disperses and ages. In this study, the sampling of gas phase organic emissions from the BnM fire was performed directly after ignition, whilst the traditional fire sampling was performed 40 min after the fire was ignited, which should have led to more optimised combustion conditions. However, the higher relative proportion of unsaturated hydrocarbons, aldehydes, PAHs and PAH derivatives in the traditional fire emissions suggest that this was not the case and that combustion of the fuels was incomplete (Fig. 9), whilst the higher proportion of saturated and oxidised products (alcohols and ketones) in the case of the BnM fire suggest better combustion conditions.

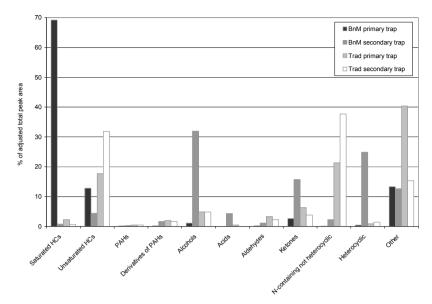


Figure 9: Comparison of the relative composition of gas phase organic compound emissions from fires lit by the BnM and traditional methods, based on the percentage contribution of compound classes to the total adjusted peak area. Results for both the primary and secondary traps are included. No filter was used for the traditional fire sampling.

5 Conclusion

There are numerous inter-related parameters which may impact on the results obtained in such a combustion study. The results reported here should be considered as preliminary, as further controlled experiments are required to fully elucidate the impacts of each variable on the resulting emissions. The results do, however, show that changes in combustion conditions have a marked effect on the organic and particulate emissions and the use of SEM and TD-GCxGC-MS in monitoring these changes has been demonstrated.

The BnM method of lighting a fire appeared to provide more optimised combustion conditions and produced a lower proportion of gas phase organic compounds which arise from incomplete combustion, including PAHs, compared to the traditional method. This is in agreement with visual observations of lower smoke production from BnM fires and thus the governmental campaign to promote this method of combustion in South Africa appears to be justified.

We have also shown that it is advisable not to remain directly next to a recently lit fire, in order to reduce exposure of people to potentially harmful air pollutants associated with emitted particles.



An integrated approach to reduce potential exposure to biomass fuel emissions should be explored, including energy efficient housing design; the provision of cost effective, energy efficient combustion appliances; as well as appropriate community education campaigns. Overall, improved household fire combustion methods and practices in the developing world have the potential to improve human health, as well as to reduce deforestation, contributions to climate change and emissions of greenhouse species such as black carbon.

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References

- [1] Statistics South Africa, Basic Results for Municipalities and Provinces, *Community Survey 2007*, Online. <u>http://www.statssa.gov.za/community_new/content.asp</u>, 2009.
- [2] Gustafson, P., Östman, C. & Sällsten, G., Indoor levels of polycyclic aromatic hydrocarbons in homes with or without wood burning for heating, *Environmental Science and Technology*, **42** (14), pp. 5074-5080, 2008.
- [3] Laumbach, R.J. & Kipen, H.M., Respiratory health effects of air pollution: Update on biomass smoke and traffic pollution, *Clinical Reviews in Allergy and Immunology*, **129** (1), pp 3-11, 2012.
- [4] Engelbrecht, J.P., Swanepoel, L., Chow, J.C., Watson, J.G. & Egami, R.T., The comparison of source contributions from residential coal and lowsmoke fuels, using CMB modelling, in South Africa, *Environmental Science & Policy*, 5, pp. 157-167, 2002.
- [5] Ortner, E.K. & Rohwer, E.R., Trace analysis of semi-volatile organic air pollutants using thick film silicone rubber traps with capillary gas chromatography, *Journal of High Resolution Chromatography*, **19**, pp. 339-344, 1996.
- [6] Le Roux, L.J., Zunckel, M. & McCormick, S.G., Laboratory controlled quantitative information about reduction in air pollution using "Basa njengo Magogo" methodology and applicability to low-smoke fuels, CSIR Report ENV-D-C-2005-004, 2004.
- [7] Forbes, P.B.C. & Rohwer, E.R., Application of laser induced fluorescence to monitor atmospheric polycyclic aromatic hydrocarbons, *Air Pollution XVIII, WIT Transactions on Ecology and the Environment*, **136**, pp. 367-377, ISSN 1743-3541, 2010.
- [8] Forbes, P.B.C., Karg, E.W., Zimmermann, R. & Rohwer, E.R., The use of multi-channel silicone rubber traps as denuders for polycyclic aromatic



hydrocarbons, Analytica Chimica Acta, doi:10.1016/j.aca.2011.11.013, 2011.

- [9] Chen, Y., Shah, N., Braun, A., Huggins, F.E. & Huffman, G.P., Electron microscopy investigation of carbonaceous particulate matter generated by combustion of fossil fuels, *Energy & Fuels*, **19**, pp. 1644-1651, 2005.
- [10] Chen, Y., Bi, X., Mai, B., Sheng, G. & Fu, J., Emission characterization of particulate/gaseous phases and size association for polycyclic aromatic hydrocarbons from residential coal combustion, *Fuel*, 83, pp. 781-790, 2004.

