

Particle air pollution and its implications in the Pacific Islands

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Cover photo: Smoke from a bus in Suva

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Acknowledgements

Four years ago, I sent an e-mail to Peter Nelson and Mark Taylor, briefly describing my idea for an air quality study in Fiji. Not only did they agree to meet and discuss the idea with me, they brought their own ideas; making the study more meaningful. Further, they agreed to supervise the research. I am thankful for their support, encouragement and direction in the course of this study.

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I began this project with great excitement and expected that it would soon settle into a routine. That didn't really happen; the whole experience has been a big adventure that I have enjoyed immensely.

Declaration

I certify that this work in this thesis has not been submitted previously, in whole or in part, for any degree at this or any other university. Nor does it contain, to the best of my knowledge, any material published or written by another person, except where acknowledged. This thesis is comprised solely of my own work.



Cynthia Faye Isley

1st August 2018

Ethics approval

Official approval to conduct this research in Fiji was granted on the 28th of April 2014, reference RA20/14 by Parmeshwar Mohan, Permanent Secretary for Education, National Heritage, Culture and Arts, Fiji and, on 12th March 2014, reference 12/2/1, by Eleni Tokaduadua, Ministry of Local Government, Urban Development, Housing and Environment, Fiji. Ethics approval was granted for this research by the Human Ethics Secretariat at Macquarie University (reference number 5201400557), dated 24th of June 2014. Approval letters are included as Appendix A of this thesis.

Abstract

Emissions arising from diesel combustion and the open burning of wastes are of concern across the Pacific Islands. Using Suva, Fiji as a case study, particulate air quality was investigated from October 2014 to October 2015. Ambient particulate concentrations were determined for total, 10 μ m, 2.5 μ m (PM_{2.5}), 1 μ m and ultrafine particle (<300 nm) size fractions at city, residential and background locations. Ion beam analysis, to determine elemental concentrations, focussed on the PM_{2.5} fraction, due to the strong evidence of adverse health impacts from PM_{2.5}. Additionally, contemporary and fossil carbon percentages were determined. Sources of PM_{2.5} were quantified by emissions inventory as well as by statistical analysis of air sampling data. Mortality risk was determined using published risk data. Indoor PM_{2.5} concentrations and sources were also investigated. Ambient PM_{2.5} concentrations (mean one year) in Suva city were close to Australian standards (8 μ g/m³). Residential area and indoor PM_{2.5} results demonstrated likely exceedance of these ambient standards. Emission inventory calculations showed diesel and waste burning to be large contributors to Suva's airborne particulate, also reflected in elevated black carbon and ultrafine particle concentrations. Statistical analysis of elemental concentrations identified diesel combustion as a major contributor to Suva's PM_{2.5} mass (21%), along with vehicle emissions (all fuel types 17%). Whilst waste burning (largely biomass) contributed only 8% to city PM_{2.5} mass, contemporary carbon concentrations indicated biomass burning contribution to airborne particulates in residential areas to be more than double compared to the city. Based on risk of black carbon exposure, mortality risk from diesel combustion in Suva (industrial, power generation and shipping) was estimated at 59 deaths per year; vehicle and open burning emissions also represented significant risk. Reduction of emissions from these three sources would improve air quality and reduce health risk. This applies for Suva and more broadly across the Pacific Islands region.

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Notes regarding thesis format and author contributions to publications

This thesis is divided into four chapters. The studies contained within this thesis are presented in Chapters Two and Three. Chapter Two quantifies the pollutant concentrations of fine and ultrafine particles in Suva's air. Chapter Three identifies the sources of fine particles in Suva's air, through the use of emissions inventory, receptor source modelling and radiocarbon methods. Chapter Three also explores the implications of these results for health risk and policy. These two chapters provide a detailed assessment of air quality, emissions sources and implications for Suva.

Chapter Two: Pollutant Concentration

Paper One

Isley C.F., Nelson P.F., Taylor M.P., Mani F.S., Maata M., Atanacio A.J., Stelcer E. and Cohen D. D. 2017. PM_{2.5} and aerosol black carbon in Suva, Fiji. *Atmospheric Environment*, 150:55-66.

Contribution

Conception and writing 100%; data analysis 100%, field work 50%, laboratory analysis 50%. Editing and Review were performed by myself, Nelson, Taylor and Cohen. Fieldwork was conducted by myself, Mani and Maata. Laboratory analysis was conducted by myself, Atanacio and Stelcer.

Paper Two

Isley C.F., Nelson P. F., Taylor M.P., Mazaheri M., Morawska L. Morrison, A.L. Atanacio A., Stelcer E. and Cohen D. D. 2017. Airborne ultrafine particles in a Pacific Island Country: characteristics, sources and implication on human exposure. *Environmental Pollution*, 231:367-378.

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Conception 50%, writing 100%, field work 100%, data analysis 95%, laboratory analysis 60%. The concept for this paper was developed with Morawska, it was written by myself. Editing and Review were done by myself, Mazaheri, Morawska, Nelson, Taylor, Morrison and Cohen. Fieldwork was conducted by myself. Data analysis was conducted by myself and Mazaheri. Laboratory analysis was conducted by myself, Atanacio and Stelcer.

Chapter Three: Source Contributions

Paper Three

Isley C.F., Nelson P.F., Taylor M.P. 2016. Managing air quality in Suva, Fiji *European Journal of Sustainable Development*, 5(4): 242-254

Contribution

Conception 100%, writing 95%, data analysis 100%. This paper was initiated and written by myself, with development by myself and Nelson. Nelson and Taylor performed editing and review.

Paper Four

Isley C. F., Nelson P. F., Taylor M.P. Atanacio A., Stelcer E., Cohen D. D. Mani F.S. and Maata M. 2018. Reducing mortality risk by targeting specific air pollution sources: Suva, Fiji. *Science of the Total Environment* 612C:450-461.

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Conception 90%, writing 100%, data analysis 80%, laboratory analysis 50%, field work 50%.

This paper was initiated and written by myself, with development by myself and Taylor.

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Paper Five

Isley C.F., Nelson P.F., Taylor M.P., Williams A.A. and Jacobsen G. 2018. Radiocarbon determination of fossil and contemporary carbon contribution to aerosol in the Pacific Islands. *Science of the Total Environment* 643:183-192.

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Conception 50%, writing 100%, data analysis 100%, laboratory analysis 50%, field work 100%. The concept for this paper was developed with Nelson, it was written by myself. Editing and Review were done by myself, Nelson, Taylor and Williams. Fieldwork was conducted by myself. Data analysis was conducted by myself. Laboratory analysis was conducted by myself, Williams and Jacobsen.

Paper Six

Isley C.F., Nelson P. F., Taylor M.P., Morrison, A.L. Atanacio A., Stelcer E. and Cohen D. D. 2018. Ambient air quality and indoor exposure: PM_{2.5} implications for health in Suva. *Air Quality and Climate Change* 52(1):35-43.

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Paper Seven

Isley C. F. and Taylor M. P. 2018. Air quality management in the Pacific Islands: A review of past performance and implications for the future. *Environmental Science and Policy* 84:26-33.

Contribution

Conception 100%, writing 95%. This paper was initiated and written by myself, with development by Taylor.

Acronyms and abbreviations

Al	Aluminium
ANSTO	Australian Nuclear Science and Technology Organisation
ASP	Aerosol sampling program (filter air sampler for PM _{2.5})
BC	Black carbon
Br	Bromine
Ca	Calcium
Cl	Chlorine
Co	Cobalt
Cr	Chromium
Cu	Copper
Fe	Iron
H	Hydrogen
IBA	Ion beam analysis
K	Potassium
LIPM	Laser integrating plate method
Mn	Manganese
N	Nitrogen
Na	Sodium
Ni	Nickel
P	Phosphorus
Pb	Lead
PICs	Pacific Island Countries
PM	Particulate matter (in air)

PM ₁	Particulate matter with effective aerodynamic diameter 1 µm or less
PM _{2.5}	Particulate matter with effective aerodynamic diameter 2.5 µm or less
PM ₁₀	Particulate matter with effective aerodynamic diameter 10 µm or less
PMF	Positive matrix factorisation
PNC	Particle number concentration (relates to ultrafine particles)
S	Sulphur
Si	Silicon
SPREP	Secretariat of the Pacific Regional Environment Programme
Sr	Strontium
Ti	Titanium
TSP	Total suspended particulate
USP	University of the South Pacific
V	Vanadium
WHO	World Health Organization
Zn	Zinc

Chapter One: Introduction

1.1 Particulate matter

Particulate matter is present in the air from a range of natural and human-influenced sources. Natural sources include soil and ocean aerosols (Viana *et al.* 2014; Karagulian *et al.* 2015), suspended by wind and wave activity; as well as forest fires (Liu *et al.* 2016) and volcanoes (Tam *et al.* 2016). Primary emissions from human activities include vehicle exhaust, industrial emissions, deliberate burning of biomass and fossil fuels as well as suspension of dust by vehicle movements and by industrial and agricultural activities (Xing *et al.* 2013). Secondary particles, formed in the atmosphere from other particles and gasses, are also a significant particulate matter component in many locations (Huang *et al.* 2014; Cary 2016). These develop from a mixture of components, including combustion emissions (Tan *et al.* 2016) and natural sources, such as biogenic emissions (Ehn *et al.* 2014) from plants.

Health implications

Particulate matter is the fraction of air pollution that is most reliably associated with human disease (Anderson *et al.* 2012). There is evidence that particulate matter is generally harmful to health; Dominici *et al.* (2014) report associations between total suspended particulates (TSP) and mortality. Similarly, a review by Anderson *et al.* (2012) shows that TSP, PM₁₀ (particulate matter < 10 µm), PM_{2.5} (particulate matter < 2.5 µm) and smoke components of particulate matter all relate to mortality, as well as to other health impacts. Toxicity studies are, however, complicated by differing particle sizes and sources at different sites, with potential for differential toxicity of particulate matter components (Osornio-Vargas *et al.* 2003; Bell *et al.* 2014; Dai *et al.* 2014; Chung *et al.* 2015; Loxham 2015; Jia *et al.* 2017). There is evidence that particles from combustion sources, including vehicle exhaust, may be more toxic (Mauderly *et al.* 2014; Hime *et al.* 2015; Kioumourtzoglou *et al.* 2015; Pun *et al.*

2015) than many natural sources such as ocean aerosols; due to both size and composition of particles; this is discussed in detail in Paper Four.

Inhaled particulate matter

Particles of different sizes penetrate the human body to differing extents. Figure 1.1, adapted from Haberzettl *et al.* (2014), summarises the fate of inhaled particulate matter. Particles 2.5 μm and smaller penetrate more deeply into the human body and are more likely to accumulate and cause damage. Evidence for a causal relationship (Brook *et al.* 2013) between exposure and mortality or adverse health effects is strongest for the $\text{PM}_{2.5}$ fraction (Choi *et al.* 2004; Franklin *et al.* 2007). In their review of health studies, Brook *et al.* (2013) conclude that exposure to $\text{PM}_{2.5}$ over a few hours to weeks can trigger cardiovascular disease-related mortality and nonfatal events; exposure over a few years increases this risk and reduces life expectancy. Discussion of the health impacts of $\text{PM}_{2.5}$ is included in Paper One. A detailed literature review of health effects studies as they relate to different sources and components of $\text{PM}_{2.5}$ is included in Paper Four. This includes discussion on the toxicity of black carbon, for which a large volume of evidence exists (Hoek *et al.* 2013).

Modifiable exposure

Whilst there are natural PM sources that are unavoidable, reductions in morbidity and mortality can be achieved by reducing modifiable PM exposure (Anderson *et al.* 2012; Brook *et al.* 2013). Indeed, reduction in $\text{PM}_{2.5}$ exposure has been demonstrated to result in improved health and mortality outcomes (Pope *et al.* 2009). Epidemiological studies have demonstrated associations between exposure to ambient air pollution and a range of morbidity endpoints (Xing *et al.* 2016; Ibrahimou *et al.* 2017; Song *et al.* 2017). Investigation of possible $\text{PM}_{2.5}$ reduction is therefore important from a public health perspective. For this reason, $\text{PM}_{2.5}$ was chosen as the main air pollutant of study for this thesis. Black carbon concentrations and sources in $\text{PM}_{2.5}$ have also been addressed throughout this study.

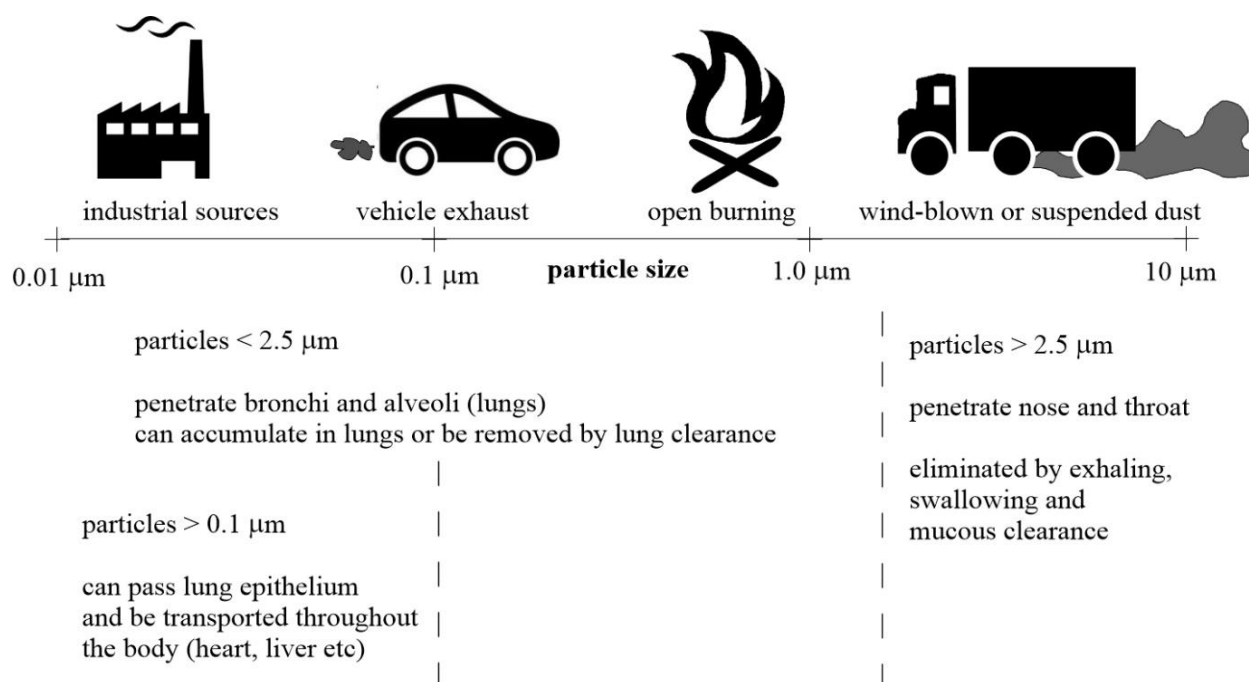


Figure 1.1 Particles of different sizes penetrate the human body to differing extents. Larger sized particles are typically associated with dust suspension, smaller particles with combustion processes. Adapted from (Haberzettl *et al.* 2014).

Ultrafine particles

If $\text{PM}_{2.5}$ are more toxic than larger fractions due to their enhanced transport within the human body, it follows that ultrafine particles (smaller than $0.1 \mu\text{m}$), may be of even greater concern (Chen *et al.* 2016). Having a large surface area per unit mass and being comparable in size to the cellular structure of the lungs (Loxham *et al.* 2015); these particles are transported to the brain and other organs (Frampton and Rich 2016). They are potentially harmful; particularly when composed of black carbon (Donaldson *et al.* 2001), diesel exhaust particles (Srivastava and Yadav 2016) or organic compounds (Akhtar *et al.* 2014; Ostro *et al.* 2015). Ultrafine particles have been related to adverse cardiac (Holland *et al.* 2017), vascular (Karotki *et al.* 2015) and lung function (Samoli *et al.* 2016) effects, markers of inflammation and diabetes (Karotki *et al.* 2014) and to brain function impairment (Allen *et al.* 2014; Cheng *et al.* 2016; Solaimani *et al.* 2017). Given the interest surrounding ultrafine particles, a monitoring

campaign to measure levels of Ultrafine Particles in Suva was also included as part of this study (Paper Two).

1.2 Pacific Islands

Fiji's location, in a global sense, is shown in Figure 1.2. The Pacific Island Countries (PICs, Figure 1.3) comprise 25 nations and territories, encompassing more than 25,000 islands of the Western and Central Pacific Ocean (Costa and Sharp 2011). There are 10 million people living in the Pacific region, on islands scattered across 180 million square kilometres (World Health Organization 2016). Whilst cultural and language diversity is great across these islands (Lal and Fortune 2000; Mühlhäusler 2002), they share a number of things in common. Whilst PICs are spread over about 15% of the earth's surface (The World Bank 2017), the PICs are each small in land area and population size. Nauru comprises a single island, covering an area of only 21 square kilometres and is home to around 12,000 people (Nations Encyclopedia 2017). By contrast, the 33 islands of Kiribati extend over 3.5 million square kilometres of ocean (The World Bank 2017). PICs have limited natural resources, narrowly-based economies and are isolated, with large distances to major markets (The World Bank 2017).

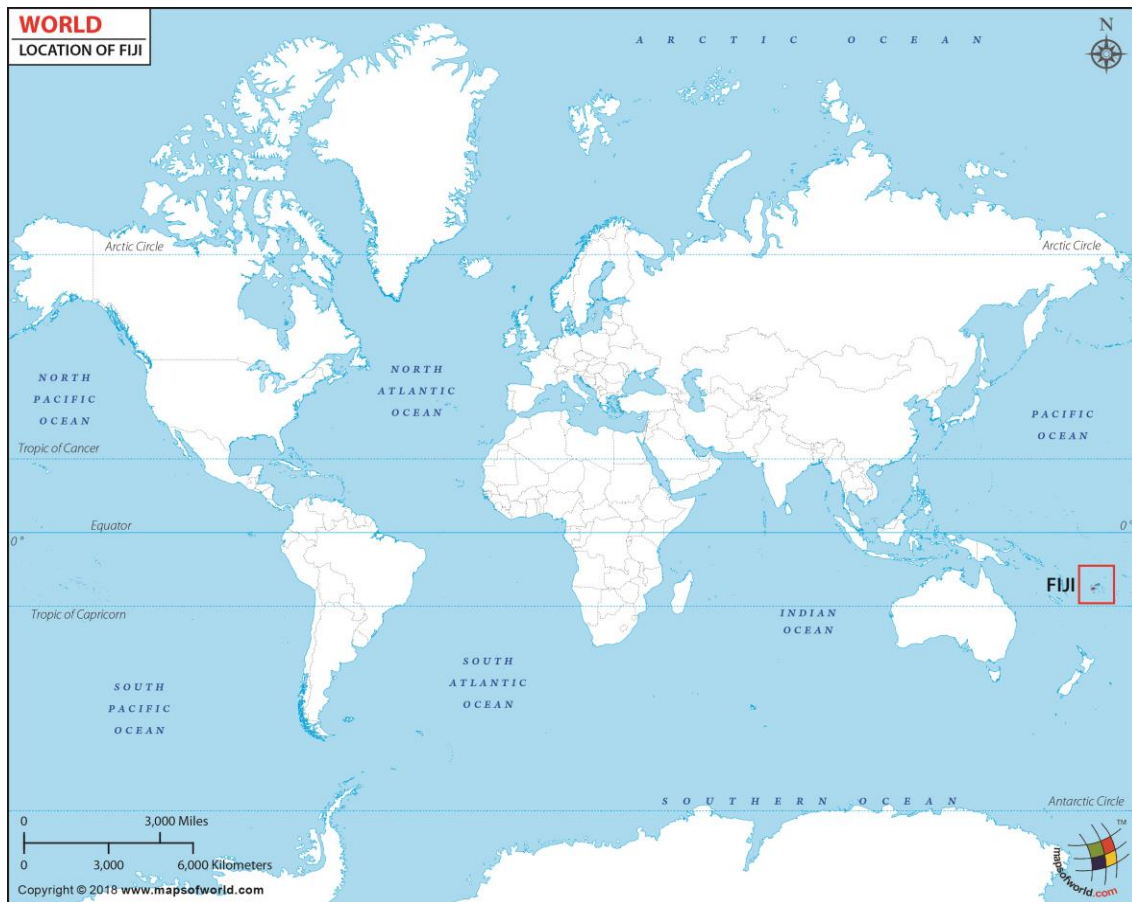


Figure 1.2 World map showing the location of Fiji. Image purchased from Map Store (order 16424).

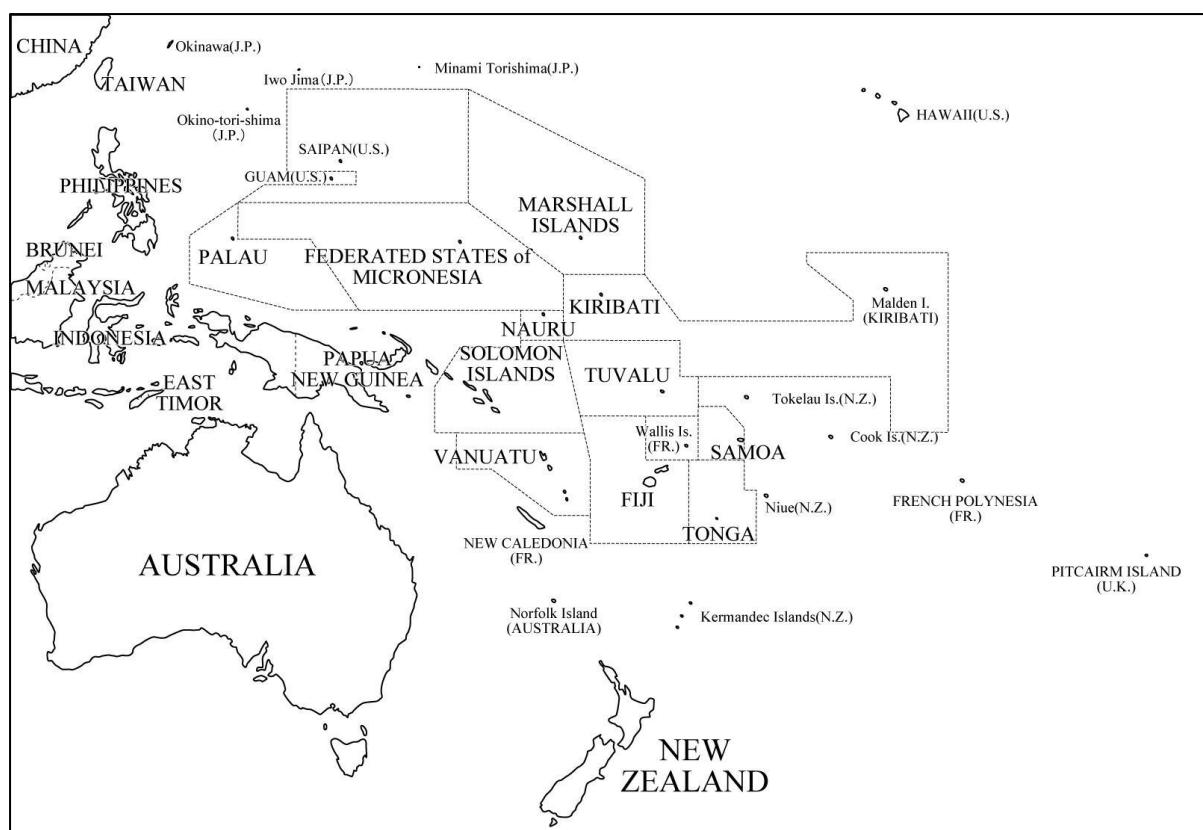


Figure 1.3 shows the Pacific Islands in more detail, including their location relative to Australia. Map source: Navarro (2014).

Waste burning

Disposal of wastes is a major challenge in PICs. Land area and hence potential sites for dumping waste are limited (situation in PICs and Hawaii (Howell 2015)); making burning of wastes an attractive option (Mataki 2011; Woodruf 2014). As population densities increase, the burning of wastes has become a health issue (Mohee *et al.* 2015). This problem also affects island communities elsewhere in the world (Marra 2016; Riquelme *et al.* 2016) and indeed cities in many developing countries (Guerrero *et al.* 2013; Mitra 2014).

Diesel

A dependence on diesel burning is another common factor across PICs. Many PICs have made use of renewable energy resources, yet diesel remains to be highly used as an energy

source for the Pacific Islands (Betzold 2016; Taibi *et al.* 2016; Lucas *et al.* 2017). This is the case even in Fiji (Dornan and Jotzo 2012), which has significant supply of hydroelectric power. Diesel generators are particularly valuable for remote PIC communities (Dornan 2014). As with waste burning, this problem is not unique to the Pacific region, but also afflicts other island communities globally (Telesford and Strachan 2017). Discussion of the usage and quality of diesel fuels in Fiji is included in Paper Three.

Air quality studies to date

Studies on air particulates in the PICs are limited. New Caledonia (primarily Noumea) introduced monitoring of PM_{2.5} in July 2016 (Association de Surveillance Calédonienne de la Qualité de l'Air 2017a). Monitoring of PM₁₀ in Noumea (Association de Surveillance Calédonienne de la Qualité de l'Air 2017b) has included chemical analysis of samples and analysis of wind directions, implicating the Doniambo industrial facility to be a significant air pollution source, along with traffic, construction and open burning sources (Escoffier *et al.* 2016). These studies have looked at particulate source contributions in a qualitative sense, rather than quantifying emissions or atmospheric loadings from different sources. Similarly, a study of total particulate (TSP) in Fiji (Garimella and Deo 2007) considered particulate sources by comparing elemental concentrations in air to those in the earth's crust. Apart from these, the other particle air quality studies available for the Pacific Islands are focussed on coastal aerosols in Samoa (Arimoto *et al.* 1987; Savoie and Prospero 1996).

This study therefore represents the most detailed study of air pollutants in the PICs. These locations have potentially significant health impacts due to diesel burning and domestic burning of biomass and waste, which to date have not been investigated. The lack of representation of this region in scientific air quality literature was a primary motivation for this study.

Fiji

Suva is located on Fiji's largest island, Viti Levu (as shown in Figure 1.4). Whilst Suva is the capital city of Fiji and the largest city in the Pacific Island Countries, its dependence on diesel for transport, power and industry and the widespread burning of wastes (both discussed in Paper Three) are common with other areas of the Pacific Islands. The findings for Suva therefore have relevance to other locations with similar climates and similar emissions practices. Suva's isolation from other population centres made it an ideal site for study, with minimal emissions interference from neighbouring areas. This meant that source contributions could be more clearly defined. Suva is also a relatively small city in terms of geographic size, at around 4 km across the peninsula and extending 15 km inland.



Figure 1.4 Fiji showing the location of Suva. Fiji consists of 332 islands and 522 islets. Of these, 106 islands are inhabited. Image purchased from Map Store (order 16424).

Air quality criteria

The national ambient air quality standards for Fiji are included within Environment Management (waste disposal and recycling) Regulations (Government of Fiji 2007a). These include threshold concentrations for PM_{10} ($50 \mu\text{g}/\text{m}^3$ as 24-hour mean) yet do not address $PM_{2.5}$. Emission standards are also given for specific particulate components such as individual metals and soot (soot not to exceed $0.01 \text{ mg}/\text{m}^3$ in emissions (Department of

Environment Fiji 2007; Government of Fiji 2007a). In the absence of compliance monitoring (Government of Fiji 2007b), these criteria do not carry great meaning. For Noumea, monitoring and air quality goals are based on European regulations, although these are not directly applicable in New Caledonia (Association de Surveillance Calédonienne de la Qualité de l'Air 2017c).

The World Health Organization (2006) has recommended a $PM_{2.5}$ goal of $10 \mu g/m^3$ in ambient air as an annual average, with a daily maximum level of $25 \mu g/m^3$. For Australia, the ambient air quality standard for $PM_{2.5}$ is $8 \mu g/m^3$ as an annual average (Department of the Environment Australia 2016), with a daily maximum level of $25 \mu g/m^3$. Epidemiological evidence contends that there is no evidence of a threshold concentration for $PM_{2.5}$, below which no adverse impacts occur (Pope and Dockery 2006). Considering the health impacts of $PM_{2.5}$ exposure, this means that any actions that reduce ambient $PM_{2.5}$ concentrations will bring health benefits to the exposed population. Measuring ambient concentrations of $PM_{2.5}$ and determining the main emissions contributing to these is therefore of value. This will allow the largest emissions sources to be targeted and improvements to be measured quantitatively.

1.3 Aims

Whilst there are concerns about the air quality in the rapidly growing cities of the Pacific Islands, very little data are available. This thesis seeks to understand the air quality conditions in the PICs, focussing on Suva, Fiji, in order to determine the best course of action for air quality management in the region.

The aims of this thesis, addressed in the studies presented in the following chapters, include:

- Measurement of the concentration of fine airborne particulates in Suva, ambient and indoor.

- Exploration of the components of Suva's fine particulates, initially in terms of black carbon content and ultrafine particle components and also in terms of elemental composition.
- Quantification of known emissions to Suva's air and identification of gaps in knowledge.
- Apportionment of Suva's fine particulates to both natural and anthropogenic emission sources.
- Assessment of the health impacts of particulate air pollutants.
- Provide the relevant government departments and other organisations with the data required to bring about policy change and effectively manage air quality.

1.4 Materials and Methods

The aims of this thesis have been achieved through the use of multiple methods. This approach provides a comprehensive and diverse set of information from which a more complete picture of air quality and its implications can be gained. Specific details of the materials and methods used for each part of the thesis are provided within the publication for each study. The interconnection of these methods is summarised in Figure 1.5.

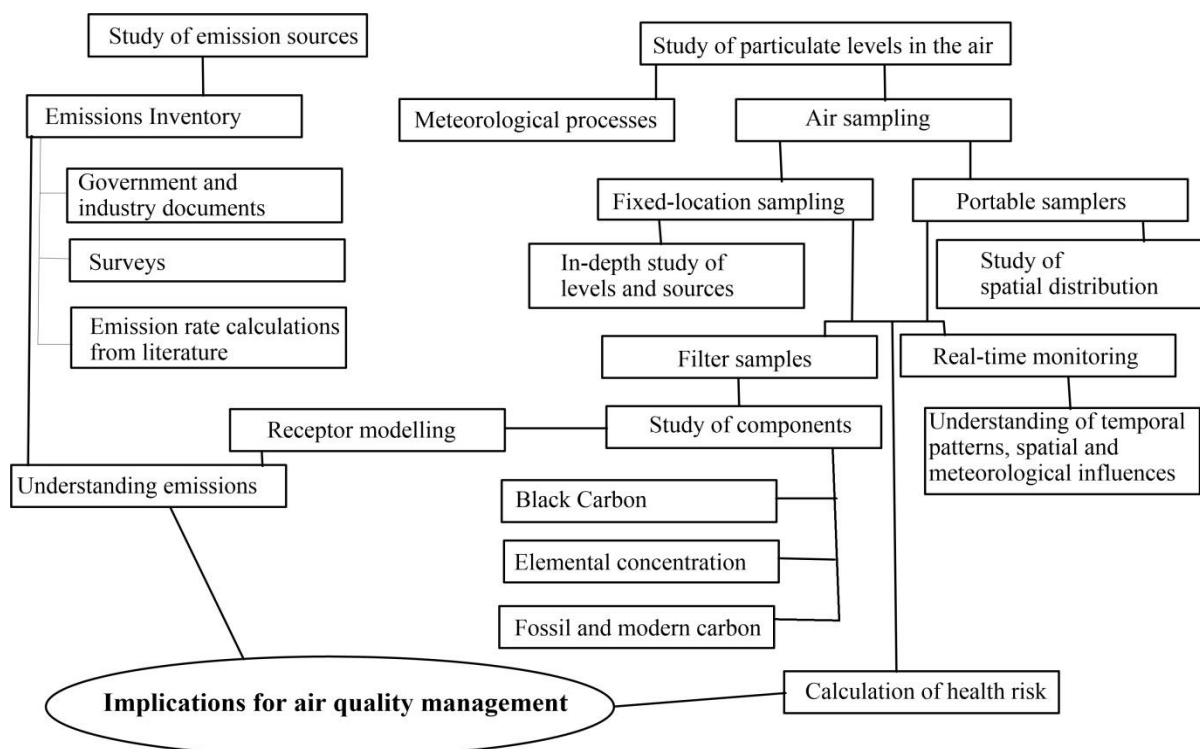


Figure 1.5 Methods used to investigate particle air pollution and its implications in Suva

In order to determine the way forward for air quality management, the sources of air emissions must be quantified in order that measurable reduction of these emission sources can be achieved. Due to the complexity of atmospheric processes, however, emissions do not provide an accurate measure of pollutant exposures (Hibberd *et al.* 2013). Therefore, the concentration of pollutants in the air must also be studied. Doing so allows calculation of the potential health risks and determination of priorities for air quality improvement. Therefore the methods in this study have followed two basic approaches, the first focussing on emissions calculation and the second on measurement of particle concentrations in air.

Emissions inventory

Using published emissions calculation guides, emissions of PM_{2.5} and black carbon from major activities were able to be calculated for Suva. Industries in Fiji and indeed across the Pacific, do not have strict reporting requirements for emissions (Government of Fiji 2007b).

This makes it difficult to develop an accurate emissions inventory. The potential impacts of these limitations are discussed in Paper Three. For diesel combustion, household waste burning and other household wood burning, emissions were reported as a range, giving upper and lower values. Nevertheless, sufficient data were available to determine basic emissions estimates. These include fuel usage by sector, imports, company reports, government reports and electricity generation data. Further information was collected using surveys. The resulting inventory, including calculations, is included as Paper Three. This provides a starting point for strategies aimed at emissions reduction, as well as demonstrating the areas in which further data collection is desirable.

Air sampling

Air sampling was carried out in Suva from October 2014 to October 2015. Again, this involved a mixture of methods.

Fixed sampling sites

A main site was established, in the city, on top of a four level building on the west of the Suva peninsula. This provides information on the exposures of people as they travel and work in the city. The Suva bus terminal, city markets, an industrial area and shipping port activities all lie within 1 km of this City site. Due to the proximity of the port, bus terminal and industry, PM_{2.5} levels at the City site would show some influence from localised activities. This City site was chosen with the aid of the South Pacific Applied Geoscience Commission (SPC), Secretariat of the Pacific Environment Programme and Department of Environment, Fiji. Of the available locations for sampling it was agreed that this site was most suitable as it more clearly represented air quality in the city centre than other available sites. This site was considered most suitable as it also served as a meteorological monitoring station for the Australian Bureau of Meteorology, meaning that high-quality wind speed and temperature data were available for comparison to air quality results.

Equipment was operated for the full year; this allowed collection of a large volume of data for this site, over different seasons, allowing more robust statistical analysis. Sampling at this site included collection of samples on filters, which were analysed for 23 different elements. Real-time sampling was also carried out at this site, which provided insight into the change in particulate levels with time of day, day of week and changing meteorological conditions.

A further two real-time monitors were placed, for comparison, at fixed sites in Suva's residential area (Kinoya) and at Suva Point. Suva Point is located on the east of the peninsula, on the University of the South Pacific lower campus, 4 km east-southeast of the City site and is mainly characterised by winds from the ocean; hence providing data for a site less affected by emissions from the Suva city area. Kinoya is a more densely populated inland residential area, 6 km northeast from the city centre, and 1.5 km northwest of two diesel- fuelled power plants. Kinoya displays less windy conditions than the other sites, impeding the dispersal of air pollutants.

These real-time monitors measured four particulate fractions (TSP, PM₁₀, PM_{2.5} and PM₁), providing a comprehensive picture of atmospheric particulate loadings at these sites. Sampling data for these fixed sites is provided as Paper One.

Elemental concentrations determined from filter samples at the fixed site were used to determine particulate sources. This differs from the emissions inventory in that it provides a measurement of the composition of the particulate captured on each filter at this site, after it has encountered atmospheric mixing processes, atmospheric chemical reactions and dilution effects. The determination of sources is done by looking at the statistical groupings of elements that occur together in the samples, using the positive matrix factorisation method (Paatero and Tapper 1994), described in Paper Four. Effectively, this provides a different way

to look at the emission sources, and considered together with the emissions inventory, is a powerful tool for determining priority areas for air quality management.

Portable samplers

Portable samplers provide a view of Suva's air quality from a different angle. These samplers were able to be operated in a diverse range of locations, providing insight on the air quality conditions in different environments. Portable samplers were used to gain 73 filter samples inside 12 homes and businesses, during three monitoring campaigns in April-May 2015 (end of wet season) and October 2014 and 2015 (dry season) across Suva (Paper Six), providing a comparison to the main sampling site in Suva city. A portable sampler was also used to measure ultrafine particle concentrations in various microenvironments around Suva (Paper Two), providing insight into the exposure impacts of transport emissions and location. By also placing portable samplers alongside other equipment at the fixed-sampling sites for a period of time, comparison was able to be made between the different parameters measured. Where possible, sampling was repeated at the same site (or a closely located site) in multiple sampling campaigns, to capture both wet and dry seasons. Blank and duplicate samples were also collected for quality control purposes, with results detailed in Papers One, Two, Five and Six.

Laboratory methods

Laboratory analysis of filter samples was carried out at the Australian Nuclear Science and Technology Organisation (ANSTO) in Sydney.

Teflon filters were firstly subject to gravimetric measurement of particulate mass, to provide the concentration of particulates in the air. The black carbon (BC) content on filters was then determined by the laser integrating plate method (LIPM). This method was particularly useful because it was non-destructive and allowed further analysis of particulates on the filters.

Detail on gravimetric and LIPM methods are provided in Paper One. These filters were then subjected to ion beam analysis (see Paper Four), providing elemental concentrations of H, Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, Sr, Pb, and N. Combined with mass and BC, these concentrations formed the basis for statistically calculating particulate source contributions.

Additionally, quartz filters were collected from the city, Kinoya residential area and Suva Point. These were analysed for carbon content, using thermal separation methods to determine total carbon, organic carbon and elemental carbon. Where sufficient carbon was available, these carbon samples were then analysed for ^{14}C content. This provided a measure of carbon particulates from fossil sources (eg. Fossil fuel burning) and modern sources (eg. biomass burning); as detailed in Paper Five.

Health risk

As detailed in Paper Four, health risk from particulate matter and specific particulate matter components were determined using published risk data. Variation of risk on a spatial basis in Suva was explored in Paper Six. Understanding of the health risks due to particulate matter in air is of paramount importance when considering management of air quality.

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Chapter Two: Pollutant concentration

Chapter Two presents the following two papers:

Paper One: Isley C. F., Nelson P. F., Taylor M. P., Mani F. S., Maata M., Atanacio A. J., Stelcer E. and Cohen D. D. 2017. PM_{2.5} and aerosol black carbon in Suva, Fiji. *Atmospheric Environment*, 150, 55-66.

Paper Two: Isley C. F., Nelson P. F., Taylor M. P., Mazaheri M., Morawska L., Atanacio A. J., Stelcer E., Cohen D. D. and Morrison A. L. 2017. Airborne ultrafine particles in a Pacific Island Country: characteristics, sources and implication on human exposure. *Environmental Pollution*, 231, 367-378.

The two papers in Chapter Two focus on quantifying particulate air pollution in Suva, Fiji. By defining the level of fine and ultrafine particles, these papers set the scene for the remainder of the study that examines the health risks associated with the air pollutants and what management interventions are needed to mitigate such impacts. They establish that whilst Suva is a small, isolated city, ambient particle concentrations are at significant levels, both from an emissions and health perspective.

Together Chapter Two's papers identify that combustion activities (vehicles, industry and open burning) are significant sources of atmospheric particles in Suva. This provides the impetus for the continued investigations presented into combustion emissions and particle sources in the remainder of the thesis. In defining local particulate air quality and meteorological conditions, these papers also constitute a basic foundation upon which the other studies depend.

Paper One

PM_{2.5} and aerosol black carbon in Suva, Fiji

Isley C. F., Nelson P. F., Taylor M.P., Mani F.S., Maata M., Atanacio A.J. , Stelcer E. and Cohen D. D. 2017. *Atmospheric Environment*, 150, 55-66.

This study (Paper One) addresses a gap in the scientific literature: what are the ambient concentrations of fine particulate (PM_{2.5}) in the Pacific Islands (specifically Fiji)? It is vital to this thesis to first establish these particle concentrations before investigating emission sources, health impacts and possible air quality improvement. The study also addresses concentrations of particulates across different size ranges (TSP, PM₁₀, PM_{2.5} and PM₁) and how these vary across Suva, from coastal aerosol to residential areas and the city. The study design therefore enables the contribution of maritime air and anthropogenic emissions to particulate loadings to be distinguished from each other and characterised separately.

In addition, Paper One explores the impacts that island meteorology and terrain have on ambient particle concentrations. These revealed that more stable nocturnal atmospheric conditions lead to higher overnight particle concentrations (Figure 2.1). Waste burning (Figure 2.2) practices across Suva (McDowall 2005) typically occur in the evening and contribute to night-time particle concentrations. Increased vehicle and industrial activities on weekdays (Figure 2.3 and Figure 2.4) were shown to cause elevated particulate levels compared to Sundays.

Meteorological impacts were modelled using The Commonwealth Scientific and Industrial Research Organisation's *The Air Pollution Model* (TAPM (Hurley 2008)) and National Oceanic and Atmospheric Administration's *Hybrid single particle Lagrangian*

integrated trajectory model (HYSPLIT (Stein *et al.* 2015)). The Pacific Islands, including Fiji and Suva, experience predominantly east to west ‘trade winds’ (Augustinus 2004; Seed 2011), meaning that air moves quickly across the land area and atmospheric residence times are low. This indicated that higher particle concentrations in city and residential areas were strongly influenced by local anthropogenic emission sources, which therefore warranted further investigation.

Black carbon in Suva city was also measured in Paper One. High black carbon levels (seen on filters, Figure 2.5) make this study highly relevant for Fijian air policy and also relevant for many other communities in the Pacific (and elsewhere: Africa, South America) that share similar climates and similar burning practices.



Figure 2.1 Looking over Suva's Walu Bay at 06:00 (October 2014). This photograph demonstrates the stable atmospheric conditions that frequently occur overnight in Suva (Paper One). Here a low-hanging layer of smoke is visible over the industrial area and residential area (in the rear of the photographs).



Figure 2.2 Waste Burning. Top: A supermarket in Suva's city centre has been burning waste in a vacant lot. The cloudy conditions shown here are typical for Suva (Paper One).

Nearby (bottom), garden wastes and rubbish are smoldering on a residential street in Suva.

Images dated October 2015.



Figure 2.3 Suva bus terminal April 2015 (rear) including the port loading area (front). Several rows of buses are lined up at Suva's bus terminal with trucks (left) for passenger transport to more outlying villages. Also shown are the Suva Markets (right, colourful roof) and shopping areas (rear).



Figure 2.4 Suva bus terminal on Sunday, April 2015. This photograph reveals a very different scene, where the bus stand is transformed into a church each Sunday. Very few buses operate on Sunday (right rear). Sunday is typically observed as a day of rest across the islands, with the reduced activity resulting in improved air quality on Sundays (Paper One).

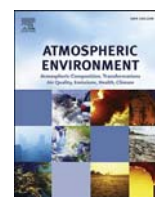


Figure 2.5 Filter samples from Suva city. The air filters each represent 24-hours sampling (30 m³ air) and show dark-coloured particles, which were found to have a high black carbon content (Paper One). Shadowing effects evident on filter rims of these early samples (October 2014) were corrected for continued sampling.



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journal homepage: www.elsevier.com/locate/atmosenvPM_{2.5} and aerosol black carbon in Suva, FijiC.F. Isley^{a,*}, P.F. Nelson^a, M.P. Taylor^a, F.S. Mani^b, M. Maata^b, A. Atanacio^c, E. Stelcer^c, D.D. Cohen^c^a Department of Environmental Sciences, Macquarie University, Sydney, NSW 2109, Australia^b Faculty of Science Technology and Environment, University of the South Pacific, Laucala Campus, Suva, Fiji^c Centre for Accelerator Science, Australian Nuclear Science and Technology Organisation, Locked Bag 2001, Kirrawee DC, NSW 2232, Australia

HIGHLIGHTS

- PM_{2.5} in Suva Fiji are generally within WHO guidelines.
- BC in PM_{2.5} are high compared to population size.
- Peak PM levels at night-time are a result of meteorological conditions.
- Back-trajectory analyses indicate low residence times of air parcels within Suva.

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ABSTRACT

Concentrations of particulate air pollution in Suva, Fiji, have been largely unknown and consequently, current strategies to reduce health risk from air pollution in Suva are not targeted effectively. This lack of air quality data is common across the Pacific Island Countries. A monitoring study, during 2014 and 2015, has characterised the fine particulate air quality in Suva, representing the most detailed study to date of fine aerosol air pollutants for the Pacific Islands; with sampling at City, Residential (Kinoya) and Back-ground (Suva Point) sites. Meteorology for Suva, as it relates to pollutant dispersion for this period of time, has also been analysed. The study design enables the contribution of maritime air and the anthropogenic emissions to be carefully distinguished from each other and separately characterised. Back trajectory calculations show that a packet of air sampled at the Suva City site has typically travelled 724 km in the 24-h prior to sampling, mainly over open ocean waters; inferring that pollutants would also be rapidly transported away from Suva. For fine particulates, Suva City reported a mid-week PM_{2.5} of $8.6 \pm 0.4 \mu\text{g}/\text{m}^3$, averaged over 13-months of gravimetric sampling. Continuous monitoring (Osiris laser photometer) suggests that some areas of Suva may experience levels exceeding the WHO PM_{2.5} guideline of $10 \mu\text{g}/\text{m}^3$, however, compared to other countries, Fiji's PM_{2.5} is low. Peak aerosol particulate levels, at all sites, were experienced at night-time, when atmospheric conditions were least favourable to dispersion of air pollutants. Suva's average ambient concentrations of black carbon in PM_{2.5}, $2.2 \pm 0.1 \mu\text{g}/\text{m}^3$, are, however, similar to those measured in much larger cities. With any given parcel of air spending only seven minutes, on average, over the land area of Suva Peninsula, these black carbon concentrations are indicative that significant combustion emissions occur within Suva. Many other communities in the Pacific Islands, as well as in Africa, Asia and South America share similar climate and similar burning practices and as such are likely to experience similar aerosol black carbon loadings. These black carbon levels indicate the need for combustion emissions, particularly those from open burning and diesel usage, to be addressed in air policy.

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1. Introduction

The public perception of the southern tropical Pacific Islands, including Fiji, is one of a clean environment. However, there is emerging evidence that substantial emissions from the combustion

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of fossil fuels, industrial and domestic waste burning are resulting in air quality that increases the risk of adverse consequences for human health. The evidentiary basis for ascribing source and atmospheric loadings of pollutants from these sources is, however, very limited for Suva, Fiji's capital city, as it is for all tropical South Pacific Island countries, which are under-represented in scientific literature. The Pacific Island Countries are 'increasingly dependent on fossil fuels' (Mishra et al., 2009), particularly diesel (Pacific Energy Summit, 2013) and, as such, are suffering greater air pollution from fossil fuel use (Mishra et al., 2009). For Fiji, diesel imports in 2010 were equivalent to 9% of the GDP (Fiji Bureau of Statistics, 2014). In many parts of the developing world, large contributions to air pollution arise from burning of wastes, including wastes associated with agriculture. In Fiji's urban areas, that have waste collection services, approximately 21% of all household waste and green waste is still burned by households (Isley et al., 2016). The same is true for Malaysia (Periathamby et al., 2009). Areas without waste collection burn a much higher proportion of their wastes (McDowall, 2005). Wiedinmyer et al. (2014) conclude, from global emissions modelling, that 'emissions of many air pollutants are significantly underestimated in current inventories because open waste burning is not included'; and that these emissions from waste burning are particularly significant in developing countries in Asia, Africa and South America. Similarly, Yevich and Logan (2003) estimate that in the world's humid tropical regions, 50% of agricultural residues are burned and that these agricultural wastes form a large proportion of biofuel combustion in Asia, Africa and Southern America which is often not counted in fuelwood usage calculations.

The need for fine particle air quality studies was highlighted at the Global Environment Facility Pacific Alliance for Sustainability Pacific Project inception meeting (Ralgavau, 2013); involving 14 Pacific Island Countries. Whilst this meeting was intended to address persistent organic pollutants from the burning of wastes, it was recognised at this meeting that fine aerosol particles from combustion also present a significant health risk. This meeting was used to refine the proposal for Suva, through consultation with Pacific Island Country representatives, in order to maximise the benefit of the study to the wider Pacific Island Community. This article presents the findings of the fine particle aerosol characterisation study for Suva.¹

Emissions from vehicles, burning of wastes and industrial sources are considered to be major contributors to air pollutant emissions in Suva; however, there is no evidence-based data delineating their relative contribution. General public complaints data indicate that air pollution is a significant community concern for Fijians (Government of Fiji, 2007). Using an analysis of complaint volumes along with visual inspections of air quality, the Department of Environment (Government of Fiji, 2007) ranked air pollution sources in order of concern: vehicle emissions, open burning of wastes at dumpsites and backyard burning of household waste, industrial emissions, agricultural burning, incinerators, cooking with open stoves, emissions from shipping vessels and dust from gravel roads. A survey investigating perceptions of air quality in Suva was carried out in 2013 (Isley, 2013), which produced a similar list of pollution sources of concern. This survey reveals that when Suva residents were asked what causes air pollution, 99% of respondents listed vehicle exhaust; 57% listed

industrial emissions, 53% open burning by households and 42% incinerators (Isley, 2013). The majority of respondents described the air pollution in Suva as being 'smoke', 'dust' or 'irritates eyes, nose or throat' (Isley, 2013). Combustion-type emissions feature heavily in these public complaints. Fine particles, namely PM_{2.5} (particles with aerodynamic diameter 2.5 µm or less), are commonly associated with emissions from combustion or secondarily formed aerosol particles, as opposed to coarse particles (aerodynamic diameter larger than 2.5 µm), commonly associated with wind-blown dust, crushing and grinding actions or re-suspension by vehicle movement (World Health Organisation, 2009). Characterisation of Suva's PM_{2.5} will provide further information about the contributions from combustion emissions.

There has been growing research interest in fine particle air pollution over recent years because of the well-established adverse human health effects from exposures, even at low concentration levels. Lelieveld et al. (2015) report that 3.3 (1.61–4.81) million premature deaths occur annually due to outdoor air pollution, mainly by PM_{2.5}. This is in accord with estimates by the World Health Organisation (2014), which also attributed around 7 million deaths in 2012 to the combination of indoor and ambient air pollution. Lim et al. (2012) ranks ambient (outdoor) PM_{2.5} air pollution as ninth on the global leading risk factors for death in 2010. Fine PM is associated with a broad spectrum of acute and chronic illness, such as lung cancer cardiopulmonary disease (World Health Organisation, 2009) and asthma (Kim, 2004). Improving air quality, in particular, reductions in PM_{2.5} concentrations, have been associated with reduced mortality risk (Laden et al., 2006; Pope et al., 2009). Considering this health-based evidence, PM_{2.5} has been selected as the main pollutant of interest in the investigation of Suva's air quality.

Black carbon (BC) aerosol particles are emitted from a range of combustion activities, with diesel engines contributing significantly to total BC emissions (McDonald et al., 2015). BC particles are very small, typically around 50 nm in diameter (Wang et al., 2015). Increased BC exposure has been associated with increased airway inflammation in adult subjects with asthma (Jansen et al., 2005) and an increased BC exposure of 1 µg/m³ relates to greater adverse health impacts than from the same increase in PM_{2.5} or PM₁₀ (Janssen et al., 2011). Similarly, in a study of six cities in the United States of America, Laden et al. (2000) found that combustion particles in PM_{2.5} from vehicles were associated with increased mortality, whereas fine particulates derived from crustal sources were not associated with increased risk. Straif et al. (2013) classified diesel engine exhaust as carcinogenic to humans, based on an association of diesel exhaust exposure with an increased risk for lung cancer. For this reason, this study also includes measurement of BC in PM.

The association of asthma and respiratory symptom occurrence in Fiji with ambient air quality is yet to be investigated. Flynn (1994b) reported current wheezing rates in Fijian children to be 21% in 1990 for children aged nine to ten years old, living in Suva. In a systematic review of worldwide variations of the prevalence of wheezing symptoms in children, Patel et al. (2008) consider the Fijian wheezing rate to be 'very high' by worldwide standards. In a ranking of the prevalence of current asthma symptoms in childhood by country (Masoli et al., 2004), Fiji was ranked 16th most prevalent out of 84 countries. Asthma has been linked to air pollution, including fine aerosol PM (Kim, 2004). Use of kerosene and solid fuels for cooking contribute to indoor aerosols (Flynn, 1994a); did not find any association between cooking fuel used and respiratory symptoms or asthma diagnosis in Suva and Nausori of Fiji. Relationship between ambient air quality and Fijian asthma rates has not been studied and further investigation is required to delineate any connections between asthma/respiratory symptoms

¹ Abbreviations used in article: ANSTO, Australian Nuclear Science and Technology Organisation; ASP, Aerosol sampling program cyclone PM_{2.5} gravimetric sampler; HYSPLIT, Hybrid single particle Lagrangian integrated trajectory model; LIPM, Laser Integrated plate method; TAPM, Commonwealth Scientific and Industrial Research Organisation air pollution model.

and air quality in Suva.

Suva, the capital city of Fiji, is located on the south-western coast of Viti Levu, the largest of the Fijian islands, as shown on Fig. 1. Prevailing winds are from the east-southeast. The terrain of the area surrounding Suva, shown in Fig. 1, rises steeply to the northwest and the west, whereas the land to the northeast is relatively flat. Along the coast to the west of Suva there are small towns, however the steep terrain inland from the coast is generally forested. The built-up-area of the city extends to the northeast for a distance of approximately 15 km. The area beyond this is a mixture of residential and agricultural landuse. Further to the east are smaller townships and villages surrounded by forested area.

If the Suva city area alone is considered, the population was 85,691 people in the most recent census (Fiji Bureau of Statistics, 2007). The adjacent metropolitan area is considered by many as part of Suva; this includes the separate local government areas of Lami (20,529), Nasinu (84,446) and Nausori (47,604), as shown on Fig. 1. The total population of the larger area was 241,270 in 2007, representing 29 per cent of the national population (Fiji Bureau of Statistics, 2007). In 2004, United Nations Habitat estimated the informal urban settlement population in Suva (squatter areas commonly referred to as ‘settlements’) to be 82,000 (UN Habitat 2004).

Fiji experiences a tropical marine climate with only slight seasonal variation in temperature. A summary of Suva's meteorology, from Fiji Bureau of Meteorology (2015) and Australian Government (2016b) is given in Table 1. Suva receives, on average, 2705 mm of rainfall per year, over 254 rain days. In terms of historical average (2000–2012), March is the wettest month, with 330 mm over 21 rain days, July is the driest month, with 124 mm over 19 rain days. March is also the warmest month with average maximum temperature of 31 °C and average minimum temperature of 24 °C. July is the coolest month with average maximum and minimum temperatures of 26 °C and 20 °C respectively. The annual average maximum and minimum temperatures for Suva are 28 °C and 22 °C respectively.

2. Method

In order to assess air quality within the city of Suva, Fiji, aerosol concentrations of PM_{2.5} were measured at three locations within the city; Suva city centre, Kinoya, and at Suva Point (Fig. 1). The period of air quality monitoring in Suva commenced 15th October 2014 and concluded 23rd October 2015. The City site, located at approximately 18 m height and 20 m elevation, on top of a four level building on the west coast of the Suva peninsula, provides information on the exposures of people as they travel and work in the city. The Suva bus terminal, city markets, an industrial area and shipping port activities all lie within 1 km of this City site. Suva Point is located on the east of the peninsula, on the University of the South Pacific lower campus, 4 km east-southeast of the City site and is mainly characterised by winds from the ocean; hence providing data for a site less affected by emissions from the Suva city area. Kinoya is a more densely populated inland residential area, 6 km northeast from the city centre, and 1.5 km northwest of two diesel-fuelled power plants. Kinoya displays less windy conditions than the other sites, impeding the dispersal of air pollutants.

Due to the proximity of the port, bus terminal and industry, PM_{2.5} levels at the City site would show some influence from localised activities. Any given location in a busy city is likely to have localised influences, however this site was selected for three main reasons. Firstly, meteorological data, collected and quality-controlled by the Australian Bureau of Meteorology (Australian Government, 2016b); is obtained from instruments affixed to the same tower (on the same building) as the air samplers for this study. Secondly, the location of this building on the western side of the city area of Suva means that air pollutant emissions from activities in the city area were likely to be detected. Thirdly, as pollutant concentration from vehicle emissions decreases with height above street level (Wang et al., 2008), the location of the sampler on top of a building (18 m height) allows some dispersion of traffic exhaust before air reaches the sampler, hence a more representative urban concentration may be determined.

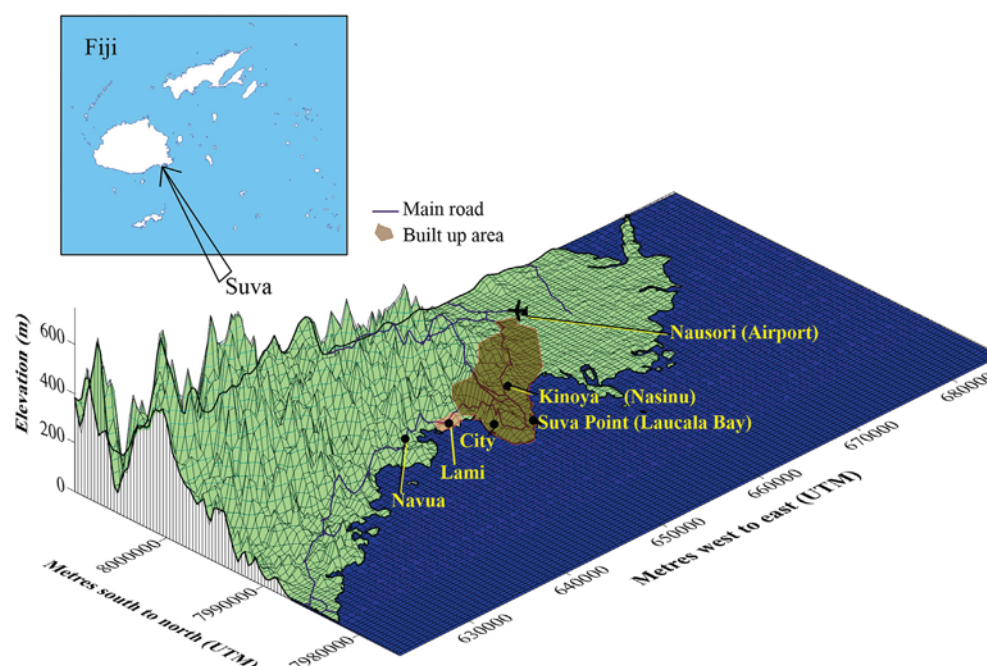


Fig. 1. Location and terrain height: Suva is located on Viti Levu, the largest of the Fijian islands, Steep terrain to the northwest and west is generally forested; land to the northeast is relatively flat and along the coast west of Suva are small towns. The built-up-area of Suva extends northeast for 15 km. Exaggerated vertical scale, elevation data source: Jarvis et al. (2016).

Table 1
Monthly air quality and meteorological summary for Suva. Fine particle air quality data (PM_{2.5}) were collected at Suva City in 2014 and 2015. Historical averages for Laucala Bay use data from 2000 to 2012. Rainfall and humidity data are from [Fiji Bureau of Meteorology \(2015\)](#). Temperature data are for Suva City, from Australian Bureau of Meteorology ([Australian Government, 2016b](#)).

	2014			2015										Avg
	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	
Monthly ASP air quality measurements ($\mu\text{g}/\text{m}^3$)														
PM _{2.5}	6.8 \pm 0.5	6.5 \pm 0.8	7.8 \pm 0.6	7.6 \pm 1.0	5.6 \pm 0.6	5.8 \pm 0.7	8.0 \pm 0.6	7.2 \pm 1.3	7.6 \pm 2.0	6.9 \pm 0.7	6.8 \pm 1.2	8.4 \pm 1.0	17.1 \pm 2.5	7.4 \pm 0.3
Suva City														
BC	1.8 \pm 0.2	1.4 \pm 0.2	2.3 \pm 0.3	2.3 \pm 0.3	2.2 \pm 0.3	2.2 \pm 0.3	3.2 \pm 0.3	2.0 \pm 0.4	2.1 \pm 0.8	1.8 \pm 0.2	1.6 \pm 0.9	2.4 \pm 0.4	4.9 \pm 0.6	2.2 \pm 0.1
Suva City														
Monthly rainfall (mm)														
Laucala Bay	218.5	59.5	579.7	292.1	273.1	188.8	85.9	126.2	55.5	48.4	132.5	127.9	98.3	175.9
Nausori Airport	225.9	97.4	735.6	312.5	311.4	172.7	152.2	150.6	75.8	69.8	138.3	170.8	122.9	201.4
Historical average	205.6	219.0	269.4	314.8	247.6	329.7	296.2	219.5	156.3	124.1	138.4	184.3	205.6	225.0
Number of rain days per month														
Laucala Bay	19	23	21	21	23	17	23	22	19	14	22	17	15	20
Historical average	21	21	24	24	21	23	23	21	20	19	19	18	21	21
Monthly average temperature ($^{\circ}\text{C}$)														
Suva City	24.9	26.0	26.5	27.4	28.3	28.2	26.8	25.1	24.1	23.5	23.0	23.6	24.9	25.6
Historical maximum	27	29	30	30	31	31	29	28	27	26	26	27	27	28
Historical minimum	21	22	23	23	23	24	23	21	21	20	20	20	21	21
Monthly average relative humidity (%)														
Laucala Bay	79	77	79	78	78	78	76	79	83	80	78	83	73	78
Nausori Airport	81	77	79	82	82	80	77	81	83	83	80	85	73	80

One Australian Nuclear Science and Technology Organisation (ANSTO) Aerosol Sampling Program (ASP) PM_{2.5} low volume cyclone sampler provides the basis for gravimetric analysis to determine PM_{2.5} concentrations at the City site. The ASP PM_{2.5} sampler used in this study was built by ANSTO according to the US EPA IMPROVE (Interagency Monitoring of Protected Visual Environments) system used across North America in their National Parks air monitoring program ([Malm et al., 1994](#); [Cohen, 1996](#); [Cohen et al., 1996](#)). Data obtained using IMPROVE samplers at three sites were found to be not significantly different to paired collocated data from US EPA Federal Reference Method samplers ([Pitchford et al., 1997](#)). Additionally, [Keywood \(1999\)](#) determined that the ASP PM_{2.5} inlet was within the performance requirements of US and Australian reference standards. PM_{2.5} data collected using ASP samplers have been widely used in recent studies ([Cohen et al., 2012, 2014](#); [Hallal et al., 2013](#); [Hibberd et al., 2013](#)); with each of these studies using identical laboratory techniques (performed at the same laboratory) for gravimetric analyses and determination of black carbon to the current study. Two Australian field studies, where PM_{2.5} from a total of six sites were sampled for 12 months each, show good correlation between Beta Attenuation Monitors (BAMs) and the ASP ([Hibberd et al., 2013, 2016](#)). A sample was collected, on a Teflon filter, each Wednesday and Sunday, for 24-h, from midnight to midnight, with a flowrate of 22 L/min. More frequent samples were collected, for approximately five weeks, during three intensive campaigns. One field blank for the ASP PM_{2.5} sampler was collected for every ten exposed samples. The mass of PM_{2.5} on the sample filters was determined gravimetrically. The filters were weighed before and after the sampling period to determine the particulate mass collected and then divided by the total volume of air that passed through the filter, to obtain the PM_{2.5} concentration. Weighing was performed under controlled temperature ($22 \pm 2^{\circ}\text{C}$) and relative humidity conditions ($50 \pm 10\%$). Using [Rasmussen et al. \(2010\)](#), this variation in laboratory humidity may potentially contribute an error of up to $\pm 0.094 \mu\text{g}/\text{m}^3$ PM_{2.5} concentration, based on average sample mass (239 μg) and volume (31.7 m³). Other studies ([Koistinen et al., 1999](#); [Buonanno et al., 2014](#)) show humidity variation in this range to have negligible effects on the mass of PM collected on Teflon filters. Calibration and quality control reports for this microbalance laboratory ([Cohen,](#)

1996), report an accuracy of $\pm 3 \mu\text{g}$ over a 12 month period; equivalent in this study to an error of $\pm 0.096 \mu\text{g}/\text{m}^3$ PM_{2.5} concentration.

Black carbon (BC) was determined, on ASP filters, using the Laser Integrated Plate Method (LIPM). Light from a HeNe laser (wavelength 633 nm) is diffused and collimated to give a uniform beam across the Teflon filter. The transmitted signal intensity is measured using a photodiode detector on each filter before and after exposure. The BC concentration is estimated from these two transmission measurements assuming a mass absorption coefficient value of $7 \text{ m}^2/\text{g}$ for carbon particles, using the method of [Taha et al. \(2007\)](#). This method provided a non-destructive estimation of BC, leaving the filters available for further chemical analyses. There is no generally accepted standard method for BC or EC and [Janssen et al. \(2012\)](#) discuss some of the difficulties with BC measurement using optical methods such as LIPM. Variability in chemical composition of aerosol at different locations and even for the same location may affect optical measurement of BC. LIPM uses a mass absorption efficiency (ϵ) to calculate BC concentration, however ϵ depends on particle size distribution and chemical composition and hence may vary over time even for the same site. In response to studies showing the high variability of ϵ for LIPM and in order to provide an improved BC calculation, calibration for the LIPM method at ANSTO was performed ([Taha et al., 2007](#)), based on experimental measurements using both test carbon and ambient aerosol samples. Reflectometer measurements with test carbon (acetylene and candle carbon) supported a mass absorption efficiency of $\epsilon = 7$; as did refractive index and density calculated from the measured elemental composition of the test carbon samples, a method similar to that used to calibrate Athelometers ([Taha et al., 2007](#)). This mass absorption efficiency value was also supported by analysis of eight years of ANSTO aerosol LIPM BC data (650 samples) from an Australian site, using experimentally determined chemical composition data for black carbon ([Taha et al., 2007](#)). Comparison to EC determined by the IMPROVE-A thermal optical method for four Australian sites ([Hibberd et al., 2016](#)), over 12 months, showed generally good agreement between the two methods, using $\epsilon = 7 \text{ m}^2/\text{g}$, however the slope of EC v BC ([Hibberd et al. \(2016\)](#) Figs. 1, 3, 4) did not pass through zero; indicating some interference from other light absorbing particles. Comparison to EC

measured by thermal desorption at two Australian sites (Hibberd et al., 2013), for 12 months, showed a good relationship (EC/BC close to 1.0) for the first seven months, however for the final five months, EC/BC > 1, attributed to carbonate interference with EC results.

Three Osiris (Turnkey Instruments Ltd.) samplers were placed in Suva; at the City, Kinoya and Suva Point. The Osiris sampler utilises the principle of low angle forward scattering of light to determine particle concentration. The Osiris samplers recorded 10-min average concentrations of total suspended particulate (TSP), particulates with equivalent aerodynamic diameter 10 µm or less (PM₁₀), PM_{2.5} and PM₁. Heated inlets were used to precondition the particle sample and account for moisture derived artefacts. The Osiris samplers use a Teflon filter, identical to that used in the ASP, with a flow rate of 0.6 L/min. These filters were weighed on a monthly basis, as per ASP filters, and these measurements were used to calibrate the continuous measurement of TSP over the 12-month sampling period. Ratio of gravimetric filter mass to mass recorded by the Osiris varied from month to month by approximately 6%. For this reason, Osiris data were considered to be approximate, however, these data are very useful for examining variation of PM_{2.5} across a typical day or week. The Osiris units at the City and Kinoya sites experienced periods of data loss due to equipment malfunction, meaning that comprehensive annual averages are not available for these sites. Available data do however indicate the differences between concentrations at each of the sampling sites. Data available are shown in Table 2.

Low-volume samplers, (SKC personal sampling pumps), with flow-rate calibrated to 3 L/min, were used to collect 27 TSP samples on quartz filters, in October 2014 and April 2015, which were also analysed gravimetrically.

A compilation of meteorological data available for Suva have been included in this study. Osiris samplers at Kinoya and Suva Point were fitted with a hemispherical cup anemometer and wind vane (Davis Instruments), to record wind speed and direction. The Australian Bureau of Meteorology (Australian Government, 2016b) monitors air temperature, wind speed and wind direction at the

Suva City air sampling site, available as hourly averaged data. Fiji Bureau of Meteorology (2015), records relative humidity and rainfall on a daily basis at Laucala Bay about 1 km from the Suva Point site and Nausori airport, 10 km northeast from the Kinoya site (Fig. 1). The Commonwealth Scientific and Industrial Research Organisation (Hurley, 2008) Air Pollution Model, (TAPM) was also used to simulate meteorology at the Suva Point and Kinoya sites, useful particularly due to the periods of missing data from the Kinoya on-site instruments. Wind direction data for the Suva Point site were also incomplete and TAPM data have been used to supplement these. In order to refine the TAPM simulations, wind speed and direction data from the Suva City site were input to the model. The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015) has been used to calculate, backwards in time, the path of air parcels arriving in Suva during the sampling period. Back trajectory calculations were carried out, at 3-hourly intervals, for the entire period of air quality monitoring; tracing the path of air parcels for a 24-h period prior to arrival at the Suva City site.

3. Results

ASP samples collected at the City site indicate ambient weekday PM_{2.5} concentrations of $8.6 \pm 0.4 \mu\text{g}/\text{m}^3$ (Table 2). Weekend concentrations were lower at $6.0 \pm 0.3 \mu\text{g}/\text{m}^3$. Black carbon accounts for approximately $30.3 \pm 1.0\%$ of PM_{2.5} in samples from Suva City and shows the same weekday and weekend pattern as PM_{2.5}; with weekday and weekend BC concentrations of $2.6 \pm 0.2 \mu\text{g}/\text{m}^3$ and $1.8 \pm 0.1 \mu\text{g}/\text{m}^3$ respectively. For Osiris results, Suva Point demonstrates lower PM_{2.5} than other sites, with levels 2.6 and 3.1 times lower those measured at the City and Kinoya, respectively. Diurnal profiles of PM_{2.5} concentration (Fig. 2) show that aerosol levels are highest overnight, between 18:00 and 05:00, with lowest levels occurring between 11:00 and 14:00. Fig. 3 shows the weekly profile for mean hourly concentration at each site, demonstrating that a similar diurnal pattern is repeated each day. The Kinoya site shows the greatest variation between morning and evening PM_{2.5}

Table 2

Summary results from all monitoring sites. Comparison of all data available for the period; PM_{2.5} measured at Suva City by ASP cyclone at 22 L/min; PM fractions (TSP, PM₁₀, P_{2.5} and PM₁) for Suva City, Kinoya (residential) and Suva Point (background); TSP by low volume samplers at or near Suva City, Kinoya and Suva Point and TSP from previous studies are shown here for comparison.

Sampler	Fraction	Date	Sample type	City	Kinoya	Suva Point
				PM concentration in $\mu\text{g}/\text{m}^3$		
ASP	PM _{2.5}	13 months Oct 2014–Oct 2015	Gravimetric 24 h n = 112 LIPM on PM _{2.5} sample n = 112	Weekday 8.6 ± 0.4	–	–
	BC in PM _{2.5}			Weekend 6.0 ± 0.3 Weekday 2.6 ± 0.15 Weekend 1.7 ± 0.12	–	–
Low-volume sampler	TSP	Oct-14	Gravimetric quartz filter 3–14 days	32.9 ± 4 n = 4	27.3 ± 4 n = 2	22.2 ± 10 n = 3
		Apr-15		33.9 ± 4 n = 4	44.7 ± 2 n = 4	26.8 ± 3 n = 10
Previous studies nearby	TSP	2000–2003 (Garimella and Deo, 2007)	Gravimetric HVAS 7 days	41 ± 4 , n = 9 Dec 2000 Suva market, 0.2 km distance	49 ± 11 , n = 4 Jul/Aug 2003 Centrepont, 1 km distance	25 ± 3 , n = 15 Jul–Nov 2003 USP campus, 1 km distance
				28.4 ± 0.2 15.2 ± 0.1 9.7 ± 0.1 3.0 ± 0.02 % contribution to total particulate measured at site	44.2 ± 0.4 21.7 ± 0.3 11.8 ± 0.1 3.6 ± 0.04 50.9 ± 0.9 22.3 ± 1.3	9.1 ± 0.1 4.8 ± 0.04 3.8 ± 0.02 1.3 ± 0.01 47.2 ± 0.9 11.0 ± 0.9
OSIRIS	TSP	City: 7 months Oct 2014–Jan 2015	Laser photometer 10 min average	28.4 ± 0.2	44.2 ± 0.4	9.1 ± 0.1
	PM ₁₀	Oct 2014–Jan 2015		15.2 ± 0.1	21.7 ± 0.3	4.8 ± 0.04
	PM _{2.5}	Apr 2015–May 2015		9.7 ± 0.1	11.8 ± 0.1	3.8 ± 0.02
	PM ₁	Oct 2015		3.0 ± 0.02	3.6 ± 0.04	1.3 ± 0.01
	TSP–PM ₁₀	Kinoya: 5 months Oct 2014–Nov 2014		46.6 ± 0.7	50.9 ± 0.9	47.2 ± 0.9
	PM ₁₀	Apr 2015–June 2015		19.1 ± 0.7	22.3 ± 1.3	11.0 ± 0.9
	–PM _{2.5}	Suva Point: 13 months				
	PM _{2.5} –PM ₁	Oct 2014–Oct 2015		23.8 ± 0.6 10.4 ± 0.6	18.7 ± 0.7 8.1 ± 1.2	27.7 ± 0.6 14.1 ± 0.6

Values in italics show the percentage contribution of particulate fraction to total particulate mass for each sampling site.

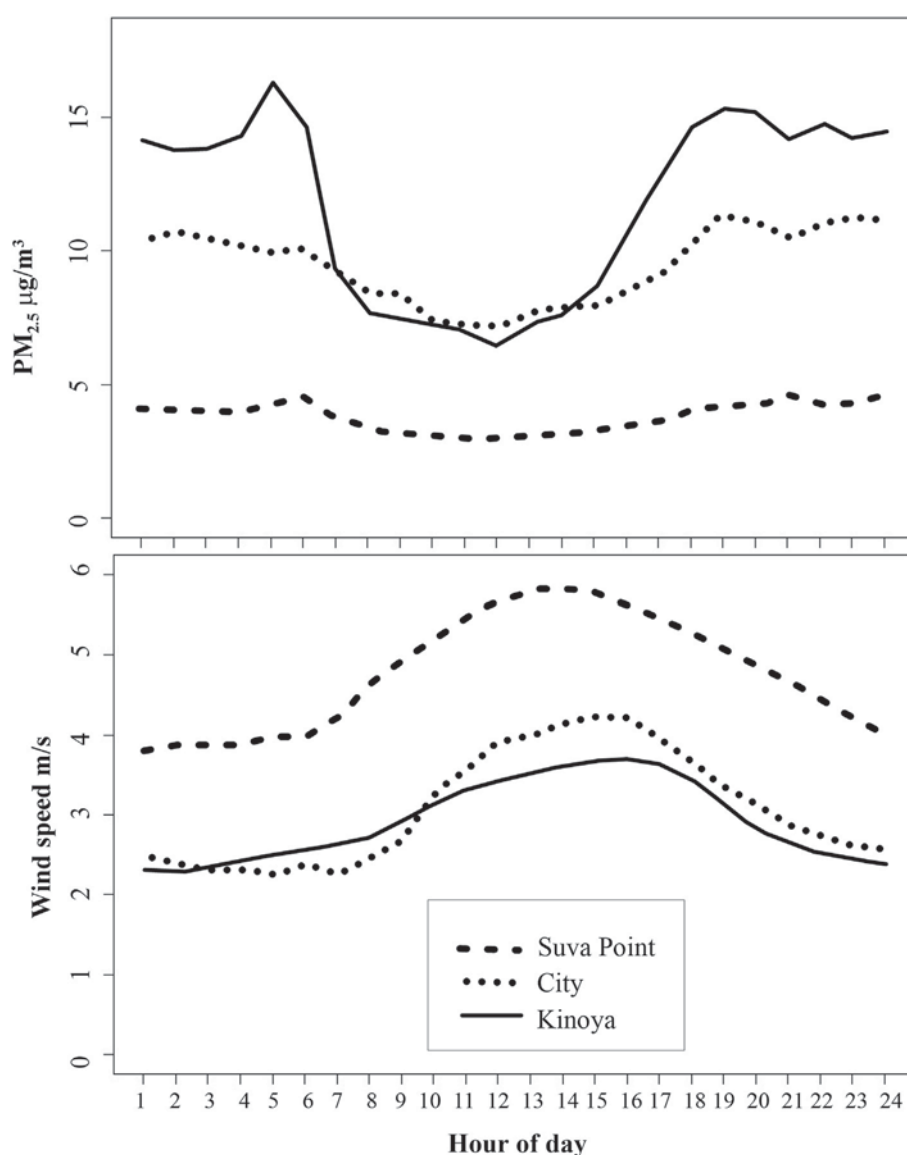


Fig. 2. PM_{2.5} and wind speed by hour of the day in Suva. Peak aerosol levels occur overnight at all sites: Suva City, Kinoya (residential) and Suva Point (background). Conversely, wind speeds are highest during the day.

concentrations. Peak PM_{2.5} concentrations ($24.2 \mu\text{g}/\text{m}^3$) occur at 03:00 on Monday mornings at Kinoya. The highest median concentrations occur at Kinoya on Wednesday evening ($23.1 \mu\text{g}/\text{m}^3$ at 21:00).

The size distribution of particulate matter recorded at each of the sampling sites is shown in Table 2. Kinoya, with TSP measured at $44 \pm 0.4 \mu\text{g}/\text{m}^3$, recorded the highest concentrations in each PM size category. On a percentage basis, across all sites, PM was measured to be comprised of 8%–14% PM₁, 19%–28% PM₁ to PM_{2.5}, 11%–22% PM_{2.5} to PM₁₀ and 47%–51% PM₁₀ to TSP. Of the three sites, Kinoya PM contained the greatest proportion of coarse matter (larger than PM_{2.5}). Quartz filter samples (Table 2) reported TSP concentrations of $25.7 \pm 5 \mu\text{g}/\text{m}^3$ for Suva Point; $24 \mu\text{g}/\text{m}^3$ (October 2014) and $48 \mu\text{g}/\text{m}^3$ (April 2015) at Kinoya and $33.4 \pm 4 \mu\text{g}/\text{m}^3$ Suva City.

Table 1 shows monthly results for PM_{2.5} measured by ASP at Suva city, alongside monthly data for rainfall, humidity and temperature, including historical data where available. Highest monthly average PM_{2.5} was recorded in October 2015 ($12.9 \pm 4 \mu\text{g}/\text{m}^3$)

and lowest monthly average PM_{2.5} was recorded in February 2015 ($5.6 \pm 1 \mu\text{g}/\text{m}^3$). For BC in PM_{2.5}, the highest monthly average was also recorded in October 2015 ($4.9 \pm 1 \mu\text{g}/\text{m}^3$) and lowest monthly average BC was recorded in November 2014 ($1.4 \pm 0.8 \mu\text{g}/\text{m}^3$). Total rainfall for Suva during the monitoring period was 2201 mm measured at Laucala Bay and 2668 mm measured at Nausori Airport, with average daily rainfall of 6 mm and 7 mm respectively.

Winds recorded by the Australian Bureau of Meteorology (Australian Government, 2016b) at Suva City during the air quality monitoring period were predominantly from the southeast (Fig. 4). Historically, winds at the Suva City site are, likewise, mainly from the southeast, however, the historical data (Fig. 4) shows a greater influence from winds in the northeastern quadrant than was observed during the monitoring period. TAPM simulated data are shown for Kinoya and Suva Point. Southeasterly winds prevailed at all three sites. On an hourly basis, for the Suva City site, highest wind speeds occurred during the day time, from 10:00 to 17:00. Winds during this period are mainly from the southeast, averaging

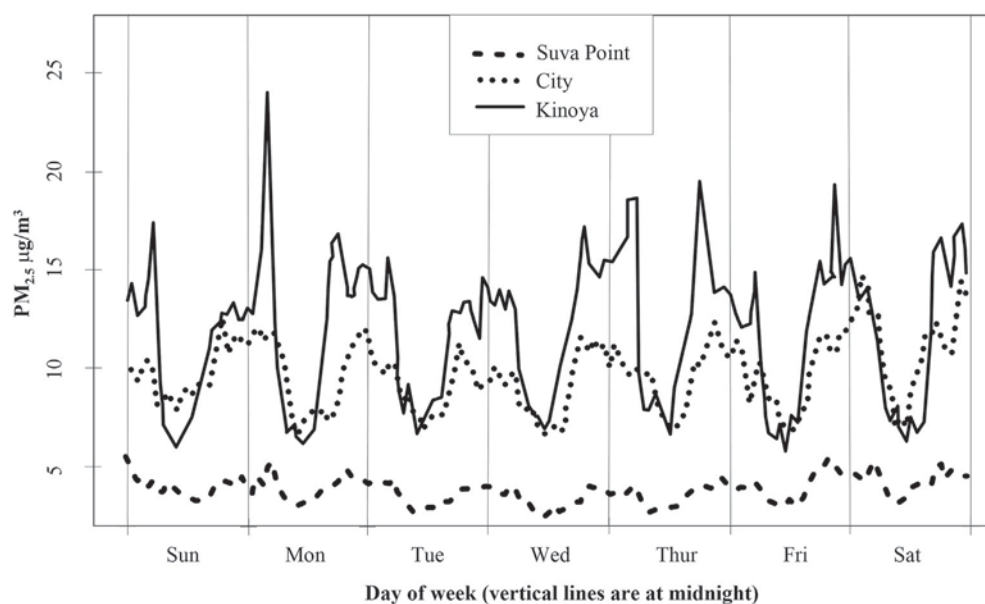


Fig. 3. Mean $PM_{2.5}$ by hour and day of the week for Suva City, Kinoya (residential) and Suva Point (background). Weekly profiles for mean hourly concentration at each site, with vertical grid-lines representing midnight; demonstrate a similar diurnal pattern being repeated each day. Greatest daily variation occurs at Kinoya, with peak $PM_{2.5}$ on Monday mornings.

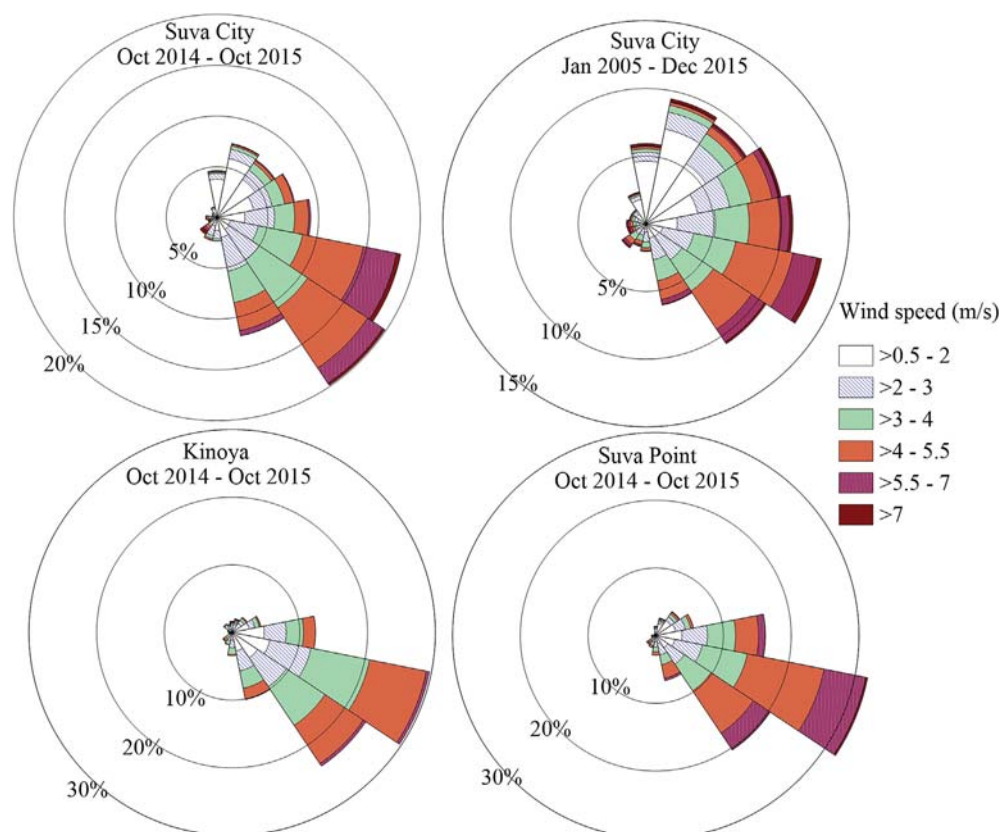


Fig. 4. Windroses for Suva for October 2014 to October 2015, showing frequency of wind speed counts for each direction. Suva City site shows Australian Bureau of Meteorology Data (Australian Government, 2016b); Kinoya and Suva Point show TAPM simulated data.

4.0 m/s and frequently exceeding 5.5 m/s (10% of the time). In the early hours of the morning, midnight to 07:00, this southeasterly component remains; however there is an increase in winds from the northeast (23% of winds) and north (14% of winds). These northerly and northeasterly winds typically have wind speeds

below 2 m/s (85% of occurrence). This same pattern of lower wind speeds at night-time is also apparent in diurnal wind speed profiles for the Kinoya and Suva Point sites (Fig. 2). TAPM wind speed has been shown for Kinoya in Fig. 2, as the anemometer at the Kinoya site recorded less than five months of data.

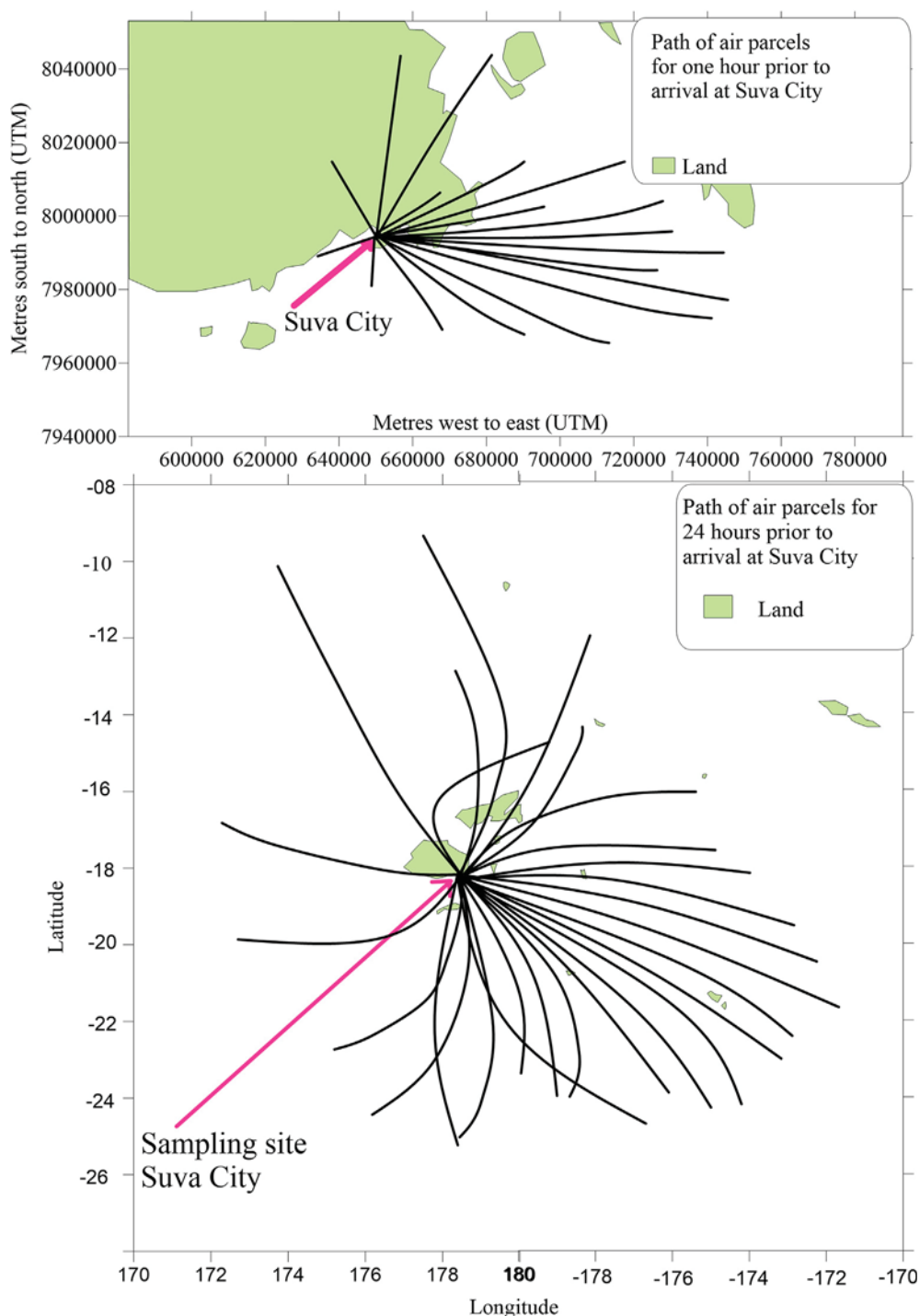


Fig. 5. HYSPLIT back trajectories for Suva City sampling site, showing prevailing south-easterly winds. A typical parcel of air travels over open ocean before arriving in Suva, with a smaller proportion of air parcels arriving from over the land area of Fiji. Air has travelled an average of 41.5 km in the hour prior to arrival in Suva, and 724 km in the past 24-h.

HYSPLIT back-trajectory analyses for one hour and 24 h prior to arrival at the Suva City site are shown in Fig. 5. When time of arrival in Suva was considered for 1-h back trajectories, it showed little difference in the origin of air parcels; with parcels of air arriving between midnight and 03:00, on average, originated from a point 3 km further north than those arriving from 09:00 to 15:00. This represents a 7.2% difference in the origins of these air parcels, considering that they have travelled an average of 41.5 km in this past hour. Typically, a packet of air sampled at the Suva City site has travelled 724 km in the 24-h prior to sampling, mainly over open

ocean waters. Air sampled at the Suva City site has typically spent only seven minutes travelling over the land area of Suva Peninsula.

4. Discussion

Meteorological conditions during the period of air quality monitoring were fairly typical of the historical averages; with some slight differences. The annual pattern of ambient air temperature during the monitoring period fits well with historic averages of maximum and minimum temperature for each month. Rainfall

during the monitoring period was, on an annual basis, typical for Suva. During December 2014, higher rainfall was experienced than the historical December average; with drier than average conditions during the period from March 2014 through to July 2015. On the whole, meteorological conditions in Suva during the monitoring period were representative of typical conditions for this location, with prevailing south-easterly winds. No statistical relationship is apparent between $PM_{2.5}$ concentrations measured and rainfall, humidity or temperature during the monitoring period. When Laucala Bay rainfall and ASP $PM_{2.5}$ levels are compared on a daily basis, $r(112) = 0.15$ and $p > 0.05$; similarly, for humidity at Laucala Bay and ASP $PM_{2.5}$, $r(112) = 0.04$ and $p > 0.05$. Also, for air temperature and $PM_{2.5}$, $p > 0.05$. The reduced incidence of winds from the northeast quadrant during the monitoring period, in comparison with historical data, may have resulted in reduced transport of air pollutants from northeastern parts of Suva to the monitoring site in the City. Indeed, from 21st to 23rd October 2015, when winds were from the northeast 73% of the time; compared to 24% over the 2014–2015 monitoring period; ASP $PM_{2.5}$ levels at the City were $13.3 \mu\text{g}/\text{m}^3$ to $21.8 \mu\text{g}/\text{m}^3$, which is well above the average $PM_{2.5}$ for the monitoring period ($7.4 \pm 0.3 \mu\text{g}/\text{m}^3$).

Continuous monitoring indicates that World Health Organisation (2006) guidelines for $PM_{2.5}$ ($10 \mu\text{g}/\text{m}^3$ as an annual mean) and Australian Government (2016a) standards ($8 \mu\text{g}/\text{m}^3$ as an annual mean) may be exceeded in some areas of Suva. A full year of data are not available for Osiris measurements at the City and Kinoya sites and hence an annual mean is not available to compare against these criteria. ASP gravimetric results for $PM_{2.5}$ at the city site indicate that weekday results, $8.6 \pm 0.4 \mu\text{g}/\text{m}^3$ (Table 2), considered alone, would exceed the Australian guideline, albeit only marginally. Weekend results are significantly lower (Table 2) however, and reduce this average to $7.4 \pm 0.3 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ and $2.2 \pm 0.1 \mu\text{g}/\text{m}^3$ for BC. Most (94%) weekend samples were collected on Sunday. In Suva, Sunday is a day when many businesses and industries do not operate, so it follows that emissions and ambient PM levels would be reduced. The Suva markets are closed on Sunday and public transport services are minimal, with a large portion of the city bus terminal being used for church services. It could therefore be argued that the weekday average concentration is more indicative of typical conditions in Suva City. Weekday results for Suva city provide an indication of the levels that workers and commuters would experience during the working week, when the city is more densely populated.

The World Health Organisation (2016) lists $PM_{2.5}$ and PM_{10} concentrations for 91 countries. These values are listed as a composite of city, and non-city stations for each country. If Fiji were added to the World Health Organisation list, it would be ranked 7th lowest according to $PM_{2.5}$, using gravimetric measurements. Suva's $PM_{2.5}$ concentrations are similar to those reported for New Zealand, or Canada. Suva's $PM_{2.5}$, by world standards, is low, which is not surprising for an isolated island city with a small population. For PM_{10} , Suva would be ranked 11th lowest, using an average of Osiris measurements from Kinoya (residential) and City sites, with similar PM_{10} to Luxembourg and Monaco. By comparison, the world median reported $PM_{2.5}$ concentration was $22 \mu\text{g}/\text{m}^3$ and median PM_{10} was $37 \mu\text{g}/\text{m}^3$, similar to PM concentrations in Brazil and Russia. Ambient $PM_{2.5}$ concentrations measured in Suva (ASP) comply with current air quality guidelines, however, as reduction in $PM_{2.5}$ concentration is associated with reduced mortality risk (Laden et al., 2006; Pope et al., 2009), there is still good reason to reduce these values where possible.

Particulate measurements at the three sites show relatively low levels of TSP compared to World Health Organisation (2006) guidelines of $90 \mu\text{g}/\text{m}^3$, Vrans et al. (2004) note that, for the Osiris, “due to the low sampling rate, 0.6 L/min, the sampling efficiency

for large particles decreases. What is called TSP is actually about PM_{20} and, probably, the sampling efficiency depends on wind speed”. Hence the true TSP concentration is likely to be higher than reported here. In a previous high-volume air sampling study of Suva, Garimella and Deo (2007) reported TSP concentration at the University of The South Pacific (Table 2), approximately 1 km from the Suva Point site, to be $25 \pm 3 \mu\text{g}/\text{m}^3$, much higher than the $9 \pm 0.1 \mu\text{g}/\text{m}^3$ reported by the Suva Point Osiris. Considering that the high volume air sampler (HVAS) in the Garimella and Deo study was located on the upper university campus, 1 km further inland than Suva Point, it would have been exposed to pollutant sources such as bus and car emissions and wind-blown dust; to a larger extent than the Suva Point sampler, located on the very east of the peninsula. These samples (Garimella and Deo, 2007) were collected more than ten years prior to this study and so it is also likely that air quality changes have occurred in this time. At Suva market, approximately 0.2 km from the Suva City sampling site, the TSP concentration level reported by Garimella and Deo (2007) was $41 \pm 4 \mu\text{g}/\text{m}^3$, once again higher than Osiris values at the City ($28.4 \pm 0.2 \mu\text{g}/\text{m}^3$). As well as sampling efficiency differences between the Osiris and HVAS, the City Osiris site was located at 18 m height, compared to the HVAS, which was located at ground level; meaning that highly localised emissions created by passing traffic would have more opportunity to disperse before reaching the Osiris sampler. The Osiris at Suva Point was also located at approximately 8 m height. The HVAS TSP results for Centrepont, 1 km from the Kinoya site, were $49 \pm 11 \mu\text{g}/\text{m}^3$, similar to the Kinoya Osiris average of $44 \pm 0.4 \mu\text{g}/\text{m}^3$. Both the Centrepont and Kinoya site were located close to ground level.

Low-volume quartz filter samples generally indicated concentrations in line with Osiris measurements. For the Suva Point site, low-volume quartz filter samples reported levels similar to Garimella and Deo (2007); indicating that the Osiris at this site is under-reporting TSP. This may be due to the presence of very coarse PM particles, greater than $20 \mu\text{m}$ effective aerodynamic diameter, that are not being captured by the Osiris at this site. At the remaining sites, low-volume quartz filter samples generally agreed with Osiris results. This may, therefore, point to localised differences between the sites chosen for the HVAS and Osiris studies, or it may also indicate that the low-volume quartz filter samplers experience similar sampling efficiency constraints to the Osiris monitors. The lower quartz filter result for October 2014 at Kinoya, $27.3 \pm 4 \mu\text{g}/\text{m}^3$, based on only two samples, is within the variation of daily Osiris TSP averages at this site. For Kinoya, daily average Osiris TSP concentrations range from $1 \mu\text{g}/\text{m}^3$ to $112 \mu\text{g}/\text{m}^3$ with median of $35.6 \mu\text{g}/\text{m}^3$ and standard deviation of $24 \mu\text{g}/\text{m}^3$; for October 2014, daily Osiris TSP at Kinoya ranged from $25 \mu\text{g}/\text{m}^3$ to $101 \mu\text{g}/\text{m}^3$.

Results from the Suva Point site demonstrate that the maritime aerosol arriving in Suva carries a very low PM load, hence the higher concentrations measured at the city and residential (Kinoya) sites show significant influence from land-based emissions within Suva. The Suva Point site demonstrates lower $PM_{2.5}$ than other sites, due to its location upwind of many emission sources in Suva. Windroses for Suva (Fig. 4) show south-easterly winds to prevail. Therefore air at the Suva Point site, on the east of the Suva Peninsula, would show little influence from land-based pollutant sources, as compared with sites on the western side of the peninsula. At Kinoya, mean TSP concentrations were 4.9 times higher than those measured at Suva Point, with $PM_{2.5}$ concentrations being 3.1 times higher. For the City, mean concentrations of all PM fractions were two to three times higher than at Suva Point, showing a significant increase in PM loading as air travels this short distance of 3.8 km across the land area of Suva. As well as being surrounded by residential area, where waste burning activity occurs, the higher concentrations measured at Kinoya reflect the less windy conditions

(Fig. 4) at this site, which impede dispersion of pollutants.

Suva's prevailing south-easterly wind direction is also noted in the back-trajectory calculations (Fig. 5); very little of the air sampled in Suva has come from the north or west. Analysis of trajectories shows that air parcels arriving in Suva during the middle of the night have followed very similar trajectories to those arriving at midday and at other times during the day, with only a 7.2% difference in the average point of origin (one hour before arrival of air in Suva). Average wind speed in Suva City for the monitoring period was 3.1 m/s, with only 3% calm periods (wind speed of 0.5 m/s or less). Residence times of air pollutants in Suva are small, with the typical packet of air arriving in Suva city having travelled over open ocean before arriving at the land area of Suva, then spending an average of only seven minutes travelling across the land area of Suva before arrival at the sampling site. With such short residence times over Suva, formation and accumulation of secondary aerosol particles would be limited.

Diurnal profiles of PM_{2.5} concentration for Suva show that concentrations decrease during the day and increase at night-time, with the most obvious day/night contrast occurring at Kinoya. There are many air pollutant sources, such as road traffic and daytime-operating industries, that have peak emission levels during daylight hours. The daily PM_{2.5} concentration profiles do not show these traffic or daytime emissions to be the dominant influence on PM_{2.5} concentrations. Burning of wastes has been visually observed to frequently occur in the evening and morning in Suva, which may contribute to this observed night-time PM_{2.5} peak, however, little data exist on the temporal patterns of waste burning by households in Suva. The practice of burning wastes, even where waste collection services exist, is a '*culturally entrenched behaviour*' (Mataki, 2011) in all Pacific Island Countries and so similar PM_{2.5} profiles may be anticipated for these countries. The strongest influence on this daily pattern of PM_{2.5} concentration would appear to be meteorology. This diurnal PM_{2.5} pattern appears to be related to wind speeds, which are highest during the day, facilitating rapid dispersal of pollutants; and lowest at night. For 1500 pairs of hourly-averaged PM_{2.5} (city Osiris) and wind speed (City) measurements, $p < 0.05$. Night-time winds also include a greater proportion of winds from the north and north-west than during the day, which may affect the sources of PM_{2.5} that arrive at the sampling site; however, south-easterly winds remain as the predominant wind direction at all hours.

Diurnal PM_{2.5} differences may also be described in terms of atmospheric stability. A simple and commonly-used scheme to describe atmospheric stability is that developed by Pasquill (1961). These describe the tendency of an air parcel to move vertically; and range from Class A, the most unstable, where vertical updrafts tend to develop; to class F, the most stable, where vertical updrafts tend to be suppressed. For Suva City, these stability classes have been calculated for the monitoring period and averaged to give a typical daily occurrence profile. Stability classes were calculated according to the method of Gifford (1961). Cloud cover data, required for these calculations, were obtained for Nausori Airport (Weatherspark, 2016). Whilst Nausori airport is 17 km from the Suva City site, where air temperature, wind speed and wind direction were recorded, this was the nearest station for which cloud cover data were available. The most prevalent stability class, experienced 35% of the time during the monitoring period, is 'D' class, or neutral conditions, which reflect the overcast conditions present much of the time in Suva. These D class stability conditions occur at all hours of the day and night, however they most frequently occur (36% of occurrence) from 14:00 to 19:00. The next most common is 'F' class, or stable conditions, which occur 21% of the time, always at night time, mainly (98% of occurrence) between 18:00 and 06:00. Under F class conditions, pollutants disperse slowly. Ground-level

emissions, from burning of wastes, wood-burning for cooking and from vehicles; combined with shallow mixing depths in the night and early morning, are the major reason why PM_{2.5} levels are highest overnight in Suva. Conversely, during daylight hours, the more unstable class A, B and C conditions occur for a combined total of 31% of the time, facilitating rapid dispersal of pollutants.

The Kinoya site shows an unusual peak at 03:00 on Monday mornings (Fig. 3). Data were analysed to ascertain if a one-off event may have greatly influenced average results for this hour. A similar peak occurred for the Suva Point site, on Saturday night, this was strongly influenced by high readings on the evening of 27th December 2014, from 19:00 to 24:00, where 10-min average PM_{2.5} concentrations as high as 233 µg/m³ were recorded. It is assumed that a local emission activity, such as a fire, had influenced this particular evening, however this does not represent typical Saturday night PM_{2.5} levels at Suva Point and so these hours (27th December 2014, from 19:00 to 24:00) were removed from calculations. For Kinoya, the highest 2.5% of 10-min PM_{2.5} concentrations were considered; these occurred over 36 separate days, with 94% of these occurring between 17:00 and 06:00. These highest 2.5% of readings were fairly evenly distributed across all days, with $14 \pm 5\%$ occurring each day of the week. Considering only Mondays, the top ten PM_{2.5} measurements occurred on four different Mondays, between 04:10 and 05:30. Therefore this Monday morning peak does not appear to be due to any one particular event, rather, this peak is due to a general pattern of higher levels occurring of a Monday morning. This may be due to localised activities near the monitoring site, such as a nearby resident or business that burns waste or cooks on a wood fire routinely on Monday mornings. Meteorological conditions also play a role in these peak levels at Kinoya, with the highest 2.5% of 10-min PM_{2.5} concentrations occurring mainly under wind-speed conditions of less than 1 m/s, as recorded by the on-site anemometer. Hence a combination of local emissions activities and limited atmospheric dispersion are likely to have combined to create this peak. Night-time levels are significant to exposure as, due to the warm climate, the majority of Fijian households do not close their house windows at night time; hence outdoor, ambient exposures also affect people as they sleep indoors. These indoor air pollution levels require further investigation.

Ambient black carbon concentrations measured in Suva are compared against a variety of other locations, worldwide, in Table 3. Use of differing field and laboratory analysis techniques, to report BC or EC, in different particle fractions, complicates comparison of these data. The (United States Environmental Protection Agency, 2012) discusses these differences, comparing BC and EC data, gained via a variety of techniques, for ten cities in the United States of America (USA) as well as other cities around the world. Overall the United States Environmental Protection Agency (2012) found (p. 255), that ratios of BC/EC were "*typically near 1 (BC/EC = 0.7–1.3)*". Hence it is possible to compare BC and EC, from different studies, but only in an approximate manner. Ratios of BC/EC may also vary for different combustion sources (Holder, 2015). An average of BC data from the United States Environmental Protection Agency (2012) report for the ten cities in the USA is included in Table 3. Much of the data in Table 3 come from Cohen (2011), who reported a multi-year summary (2000–2009) of black carbon concentration in PM_{2.5} measured as part of the ANSTO air sampling program. Hence these data are obtained using the same ASP samplers, Teflon filters and laboratory techniques (LIPM) as were used for Suva. These data represent an average over multiple sites for each country listed, representing mainly city areas; with the inclusion of a smaller number of sites from less densely populated districts as detailed in Cohen (2011). Further data were obtained from the European Union (EU) 'AIRBASE' database (European Environment Agency, 2014), which provided

Table 3

Suva's BC in PM_{2.5} is presented in comparison with other locations. BC and EC data are presented here for various locations. Half of these locations (Cohen, 2011), used the same techniques as the current study, representing a multi-year (2000–2009) average over multiple sites for each country, from mainly city areas; with the inclusion of a smaller number of sites from less densely populated districts. Other locations may be compared only in an approximate manner, due to differences in field and laboratory techniques.

Location	BC	PM _{2.5}	Carbon measurement
Australia urban	0.81 ± 0.02	5.56 ± 0.09	BC in PM _{2.5} (Cohen, 2011)
London urban	0.86 ± 0.02	—	EC in PM ₁₀ (Jones and Harrison, 2005)
EU rural	1.61 ± 0.90	—	Various methods (European Environment Agency, 2014)
EU kerbside	3.15 ± 0.28	—	EC in PM ₁₀ (Jones and Harrison, 2005)
London kerbside	3.38 ± 0.03	—	BC in PM _{2.5} (Cohen, 2011)
Belfast urban	1.10 ± 0.02	—	Various methods (United States Environmental Protection Agency, 2012)
New Zealand urban	1.22 ± 0.02	4.9 ± 0.2	EC in PM _{2.5} (Viana et al., 2006)
USA 10 cities BC average over 10 cities	1.26 ± 0.20	—	EC in PM _{2.5} or PM ₁₀ (European Environment Agency, 2014)
Barcelona urban	1.9 ± 0.2	25.0 ± 0.8	This study: BC in PM_{2.5}
Germany urban	2.03 ± 0.16	—	BC in PM _{2.5} (Cohen, 2011)
Suva Fiji urban	2.2 ± 0.1	7.4 ± 0.3	
Indonesia urban	2.99 ± 0.03	26.7 ± 0.1	
Korea urban	3.25 ± 0.02	15.7 ± 0.1	
Pakistan urban	3.39 ± 0.04	15.7 ± 0.1	
Malaysia urban	4.76 ± 0.01	49.8 ± 0.2	
China urban	5.08 ± 0.02	26.7 ± 1.6	

annual summaries of available air quality data throughout the EU. These data included a composite of BC and EC measurements, reported in either PM_{2.5} or PM₁₀ and are summarised into data representing rural sites ('EU rural') and data collected within 10 m of roads in highly trafficked urban areas ('EU kerbside'). Germany was the only country in the AIRBASE data set to report 'urban background' levels, shown in Table 3, representing a composite of EC in PM_{2.5} or PM₁₀ at seven sites across Cottbus, Frankfurt and Bernau (2008–2012). It is surprising that these German data, from larger, more industrialised cities, provide the closest match to BC concentrations in Suva. Suva's BC is around half of that reported for China and similarly, lower than many other highly populated urban centres in; Malaysia, Barcelona Spain, Pakistan, Korea and Indonesia; Suva's BC is also low compared with kerbside EC levels in London (Jones and Harrison, 2005) and the EU. It is noteworthy that Suva's BC levels are higher than those reported in urban USA, London England, Belfast Ireland, New Zealand and Australia. Considering the low residence time of air parcels in Suva (approximately 7 min), these BC concentrations recorded in Suva indicate that significant combustion emissions are occurring within Suva. Black carbon concentrations are significantly lower on weekends than on weekdays (Table 2). As noted above, Suva has decreased transport and industrial activity on Sundays as compared with weekdays. It would appear that these transport and industrial activities that occur with greater intensity on weekdays in Suva contribute significantly to BC concentrations.

5. Conclusion

Whilst the levels of PM_{2.5} in Suva City are within World Health Organisation guidelines, continuous monitoring suggests that the levels in residential areas of Suva may be close to or exceed these guidelines. Although the residence times of air parcels in Suva are small, black carbon concentrations in Suva are close to those of much larger cities, suggesting that combustion emissions in Suva are a significant source of particulate air pollution. These elevated black carbon concentrations warrant continued investigation; due to the variation in BC and EC results between available field and laboratory methods, an alternative measurement method is recommended in order to confirm these BC levels for Suva.

It is likely that other Pacific Island cities are similarly impacted

by combustion emissions, which require ongoing investigation. For more inland cities in the developing world, that practice similar combustion activities, yet do not have the benefit of strong maritime winds, PM_{2.5} and black carbon levels are likely to be higher than those measured in Suva. Reducing combustion emissions in Suva would reduce BC and PM_{2.5} concentrations, reducing the health risk from fine particulate exposure in Suva.

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Paper Two

Airborne ultrafine particles in a Pacific Island Country: Characteristics, sources and implications for human exposure

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Supplementary information in Appendix B of this thesis.

Paper Two extends the quantification of atmospheric particulates by introducing measurement of ultrafine particles (smaller than 100 nm).

Because their size is comparable to biological molecules, inhaled ultrafine particles are characterised by strong mobility in the human body and the ability to affect cellular function (Gramowski *et al.* 2010). This is true for all ultrafine particles, but is exacerbated by size, composition, surface structure, solubility and shape (Dhasmana *et al.* 2017). Terzano *et al.* (2010) propose pathways for ultrafine particle toxicity; including oxidative stress effects on airways, heart and the nervous system.

The concept for this paper was formed after initial sampling in Suva revealed elevated black carbon concentrations (Paper One), particularly relative to measured PM_{2.5} concentrations. It was proposed that high black carbon concentrations reflected high rates of fresh combustion emissions in Suva and yet PM_{2.5} remained fairly low (Paper One).

Meteorological modelling showed that air masses remain over the Suva Peninsula for only 7 minutes on average (Paper One), providing limited time for emitted primary particles to react and form larger particles. In the presence of sunlight, black carbon (soot) particles quickly age by developing a coating of secondary species including sulphate, nitrate,

organics and water (Moffet and Prather 2009). Whilst atmospheric reactions begin rapidly (Wiedensohler *et al.* 2009), this ‘aging’ of primary black carbon typically occurs over a period of three hours (Moffet and Prather 2009) to more than 24 hours (Van Poppel *et al.* 2005; Gentner *et al.* 2012) or three days in the case of biomass burning smoke (Ortega *et al.* 2013).

Considering Suva’s black carbon concentrations in PM_{2.5} (Paper One), ultrafine particle concentrations, particularly near emission sources (such as roads) would also be expected to be elevated. Paper Two shows that this holds true for Suva. This study further considers the relationship between ultrafine particle concentrations, PM_{2.5} and black carbon measurements at different locations across Suva.

Using a similar approach to that in Paper One, this study considers ultrafine particles as they vary with time and meteorology at the Suva city monitoring site. In addition, the equipment used (Nanotracer (Phillips 2017)) allowed for sampling on a mobile basis. The Nanotracer was calibrated against a Condensation Particle Counter (CPC), using NaCl (salt) particles as detailed in Buonanno *et al.* (2013). The Nanotracer has shown good agreement with Condensation Particle Counters, particularly when black carbon (soot) and salt (NaCl) particles were measured (Buonanno *et al.* 2014). Considering that salt (and black carbon) are prevalent in Suva’s particulate (PM_{2.5}, Paper Four), this instrument was well suited for the purpose of mobile sampling.

Both the fixed-location and mobile sampling implicated road traffic emissions in Suva as a considerable source of ultrafine particles. As a result, this study highlights that commuters, transport workers and those working alongside roadways have increased particulate exposure and face a disproportionate risk (Figures 2.6 to 2.9). Exposure is also

exacerbated in public transport vehicles that operate with open windows (Figure 2.10 to 2.11).

Whilst the study relies on a short monitoring campaign, it indicates that ultrafine particulate concentrations in Suva are at levels of concern for health. Ultrafine particle studies of similar time duration have been used elsewhere to indicate where further investigation is warranted (Mullen *et al.* 2011; Wangchuk *et al.* 2015; Cheng 2017).

The representativeness of this monitoring period, in terms of meteorology and particulate composition compared to longer-term data are discussed in section 3.1 of Paper Two (page 65 and 66). These show the typical pattern of prevailing south-easterly winds, yet with a higher proportion of night-time winds from the northeast compared to the longer study period (2014-2015) in Paper One. This higher proportion of north-easterly winds, however, is reflected in historical data averages (2005-2015, Paper One Section 4) for wind directions recorded at this site. Stability classes calculated for this period (Paper Two Section 3.1.4) were similar to those measured over the 2014-2015 monitoring period (Paper One, Section 4), particularly in terms of the typical daily occurrence profile where stable conditions dominate night-time atmospheric conditions and less stable conditions prevail during the day. This study therefore provides further evidence that combustion emissions in Pacific Island Countries require to be examined and addressed.



Figure 2.6 Roadside stalls. Stall holders in Suva city centre (2016) would be exposed to particles from vehicle exhaust throughout their workday.



Figure 2.7 Cars and trucks in Suva typically operate with windows open. The taxi driver (front), truck driver (rear) and the man on the back of the truck would all be exposed to vehicle exhaust particulates (Suva city centre, 2016).



Figure 2.8 Snack sellers at the Suva bus terminal. Many snack sellers also operate at busy road intersections, selling snacks to drivers and would be highly exposed to vehicle exhaust particulates (location – Suva, 2016).



Figure 2.9 Bus drivers and passengers are exposed to exhaust. Whilst these buses have closable windows and doors, they are typically left open due to the heat. The exhaust from buses in front/ beside enters vehicles (location – Suva, 2016).



Figure 2.10 Trucks are a popular form of passenger and goods transport in Suva. Passengers would be exposed to vehicle exhaust (location – Suva, 2016).



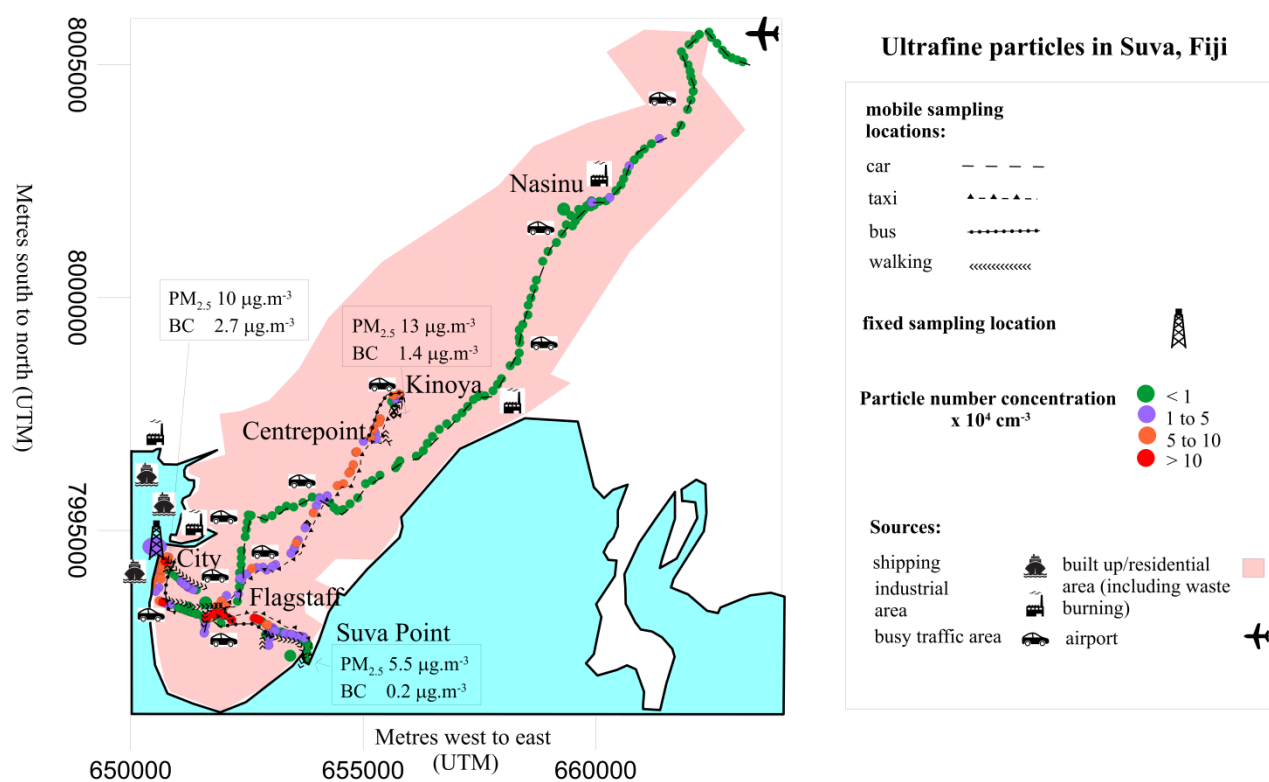
Figure 2.11 Many buses in Fiji have open windows, with a roll-down shield for rain (Suva 2015), exposing passengers to exhaust emissions.

Graphical abstract for Paper Two

Airborne ultrafine particles in a Pacific Island country: characteristics, sources and implications for human exposure

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Airborne ultrafine particles in a Pacific Island country: Characteristics, sources and implications for human exposure[☆]



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ABSTRACT

The Pacific Islands carry a perception of having clean air, yet emissions from transport and burning activities are of concern in regard to air quality and health. Ultrafine particle number concentrations (PNCs), one of the best metrics to demonstrate combustion emissions, have not been measured either in Suva or elsewhere in the Islands. This work provides insight into PNC variation across Suva and its relationship with particle mass (PM) concentration and composition. Measurements over a short monitoring campaign provide a vignette of conditions in Suva. Ambient PNCs were monitored for 8 day at a fixed location, and mobile PNC sampling for two days. These were compared with PM concentration (TSP, PM₁₀, PM_{2.5}, PM₁) and are discussed in relation to black carbon (BC) content and PM_{2.5} sources, determined from elemental concentrations; for the October 2015 period and longer-term data. Whilst Suva City PM levels remained fairly low, PM_{2.5} = 10–12 µg m⁻³, mean PNC ($1.64 \pm 0.02 \times 10^4$ cm⁻³) was high compared to global data. PNCs were greater during mobile sampling, with means of $10.3 \pm 1.4 \times 10^4$ cm⁻³ and $3.51 \pm 0.07 \times 10^4$ cm⁻³ when travelling by bus and taxi, respectively. Emissions from road vehicles, shipping, diesel and open burning were identified as PM sources for the October 2015 period. Transport related ultrafine particle emissions had a significant impact on microscale ambient concentrations, with PNCs near roads being 1.5 to 2 times higher than nearby outdoor locations and peak PNCs occurring during peak traffic times. Further data, particularly on transport and wet-season exposures, are required to confirm results. Understanding PNC in Suva will assist in formulating effective air emissions control strategies, potentially reducing population exposure across the Islands and in developing countries with similar emission characteristics.

Suva's PNC was high in comparison to global data; high exposures were related to transport and combustion emissions, which were also identified as significant PM_{2.5} sources.

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1. Introduction

Suva is the most populous city of the Pacific Island Countries; located on a peninsula, in the southeast of Viti Levu, the largest of

the Fiji Islands. With Fiji's population becoming increasingly urban, approximately 25% of Fiji's population, or 218,00 people (Government of Fiji, 2012) live in Suva. Public complaints data indicate that air pollution is a significant community concern for Fijians (Department of Environment Fiji, 2007), particularly vehicle emissions, burning of wastes and industrial emissions. Whilst PM_{2.5} concentrations in Suva generally complied with the annual average World Health Organisation guideline of 10 µg/m³ (World Health Organisation, 2006; Isley et al., 2017b); black carbon (BC) concentrations in Suva City were similar to much larger, more industrialised cities, and comprised 30% of PM_{2.5} (Isley et al., 2017b). Statistical analysis of source contributions to Suva's PM_{2.5}, using

Abbreviations: ASP, aerosol sampling program PM_{2.5} sampler; BC, black carbon; CF, correction factor; dp, ultrafine particle diameter; HVAS, high volume air sampler; PM, particulate matter, subscript numbers denote size fraction in µm; PMF, positive matrix factorisation; PNC, ultrafine particle number concentration; TSP, total suspended particulate.

[☆] This paper has been recommended for acceptance by Eddy Y. Zeng.

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elemental concentration data, show that 42%–48% of PM_{2.5} originates from combustion sources: including diesel burning by small ships, industry and for power generation; open burning of wastes; road vehicle emissions and heavy fuel oil combustion by large ships (Isley et al., 2017a). Combustion emissions are a source of fine (PM_{2.5} mass concentration of particles with aerodynamic diameter < 2.5 µm) and ultrafine aerosol (diameter < 0.1 µm); and diesel emissions are typically characterised by particles in the ultrafine size range (Kittleson et al., 2004; Ning et al., 2013). The prevalence of diesel combustion emissions in Suva (Isley et al., 2016) indicates the likelihood of elevated PNCs.

Whilst many aerosol health-risk studies have focused on PM_{2.5}, ultrafine particles may present a greater health risk. In a review of the potential exposure routes and health risks of ultrafine particles, Chen et al. (2016) concluded that ultrafine particles play a major role in adverse impacts on human health and are likely responsible for many of the health impacts currently attributed to PM_{2.5}. Ultrafine particles have large surface area per unit mass and are comparable in size to the cellular structure of the lungs (Loxham et al., 2015); transporting to the brain and other organs (Frampton and Rich, 2016). This makes them potentially harmful to human health; particularly when composed of black carbon (Donaldson et al., 2001), diesel exhaust particles (Srivastava and Yadav, 2016) or organic compounds (Akhtar et al., 2014). Ostro et al. (2015) reported significant positive associations between ultrafine particle components (elemental carbon, metals and mobile sources) and ischemic heart disease. Ultrafine particles have been related to adverse cardiac (Holland et al., 2017), vascular (Karotki et al., 2015) and lung function (Samoli et al., 2016) effects, markers of inflammation and diabetes (Karotki et al., 2014) and to brain function impairment (Allen et al., 2014; Cheng et al., 2016; Solaimani et al., 2017).

Whilst very limited research has been conducted addressing air quality in the Pacific Islands, it is apparent that all Pacific Island countries share similar air quality concerns to Suva (Fiji). Monitoring of PM₁₀ has been carried out in Noumea, by L'Association de Surveillance Calédonienne de Qualité de l'Air (Escoffier et al., 2016), as well as limited PM_{2.5} analysis (Gleye, 2010). Whilst no quantitative source apportionment data are available for Noumea, the primary source implicated for episodes of elevated PM was the burning of diesel oil at the Doniambo thermal power and nickel processing plant; other elevated PM₁₀ occurrences in Noumea were attributed to road traffic, burning and construction (Escoffier et al., 2016). The dependence on diesel fuel combustion for energy is common across the Pacific Islands (Dornan and Jotzo, 2012; Keruring van Elektrotechnische Materialen te Arnhem, 2012; Pacific Energy Summit, 2013; Taibi et al., 2016). Further to this, the burning of biomass on a household level, for cooking purposes and disposal of household or agricultural wastes, is also a concern with regard to air quality across the Pacific Islands as it is in other developing countries of Asia, Africa and South America (Thaman et al., 2003; Periathamby et al., 2009; Matak, 2011; Owens et al., 2011; Wiedinmyer et al., 2014; Isley et al., 2017a). The authors are not aware of any studies conducted in Pacific Island countries investigating PNCs and, considering their potential public health implications, this is an area that requires research.

In light of the above, the aim of this paper is to address the lack of quantitative and scientific knowledge on ultrafine particle characteristics in the Pacific Islands through a case-study of Suva. This study provides a vignette of PNCs in Suva's air, both ambient levels and for various microenvironments, as well as their relationship with PM concentrations and sources. Ultimately this research seeks to demonstrate whether elevated PNCs are present, and to indicate where further investigation of PNCs may be warranted. The perception of Suva and other Pacific Island cities is that,

as the islands are small and isolated, with strong ocean winds and relatively low human populations, the PNCs would be low. This study aims to test this assumption and inform a future policy course regarding action on ultrafine particle air quality in the Pacific Islands.

2. Method

2.1. Study design

Particulate matter in terms of mass concentration of total suspended particles (TSP), PM₁₀, PM_{2.5} and PM₁; PNC and mean particle diameter (dp), were recorded at a fixed-site monitoring station: the Suva City site (see Graphical Abstract) over an eight day period, from 20th to 27th October 2015. At the City site, equipment was affixed approximately 18 m above the street level, to a tower on top of a four-level office building on the west coast of the Suva peninsula. This same site is used by the Australian Bureau of Meteorology (Australian Government, 2016) to collect hourly-averaged air temperature, wind speed and wind direction data. Data collected at this site also include gravimetric PM_{2.5} samples, for a period of one year, including the October 2015 period; analysed for BC and elemental concentration. Air particulate sources, determined using positive matrix factorisation for elemental concentrations, are also available. This location was therefore useful for interpretation of the PNC and PM concentration and characterisation of particulates in the Suva airshed. Being located on the western side of the Suva peninsula, air sampled at this site has travelled over the land and city area of Suva. The Suva bus terminal, city markets, an industrial precinct and shipping port activities all lay within 1 km of this Suva City site.

Mobile measurements of PNC and dp were collected whilst outdoors and commuting by different methods: walking, bus, taxi and private car. On the 19th and 20th October 2015, 8 h of measurements (16 s averages) were collected across Suva. The instrument used for this collection, the Nanotracer (described below), allows markers to be placed in the data by the press of a button on the instrument. Each time a marker was activated, the marker number, relevant location, activity and other relevant factors were noted in a log-book, along with the time, for later addition to the data file. The microenvironments visited are summarised in Table 1. Locations sampled include Kinoya, Centrepont, Flagstaff, Suva City and Suva Point, as well as travel in-between these locations. Kinoya is a residential suburb located in the most densely-populated council area of Fiji (Nasinu). Centrepont is a busy traffic area on the intersection of two main roads, comprising two shopping centres. Flagstaff is also a busy traffic area, with the intersection of roads from the city, Suva's northern suburbs and the University of the South Pacific to the east. Suva Point receives mainly ocean winds; although the road west of Suva Point (towards Flagstaff) is a busy thoroughfare for university traffic. Suva City is the centre of economic and transport activity, featuring the central bus terminal for the region. During mobile measurements, the Nanotracer was worn on a belt, with a sampling tube attached to the shirt collar, as to gain measurements indicative of the air in the breathing zone. During data collection, time, location and mode of transport were noted, along with whether measurements were taken near a road (compared to within a property); and whether windows of vehicles were open or closed. GPS location was also logged every 16 s; using the Greenalp real time tracker (<https://www.greenalp.com/RealTimeTracker/>) on a mobile telephone, recording spatial co-ordinates for each Nanotracer measurement and enabling mobile paths to be mapped (Graphical Abstract).

Cloud cover data were also used in order to determine atmospheric stability and mixing height. Cloud cover data for the

Table 1
Microenvironments visited during mobile PNC sampling in Suva, Fiji during October 2015.

Communing Microenvironment		Sampling time	Detail
Bus	4 bus trips	09:10	Flagstaff to Suva Pt
	Buses had open windows	14:30	Kinoya to Centrepont
		15:40	Flagstaff to city
		15:55	City
Car	2 trips by private car	20:00 to 22:30	Windows closed and air conditioning on
Taxi	Front windows open, seated in back	13:10	Flagstaff to Kinoya. Front windows open, seated in the back
		14:55	Centrepont to Flagstaff
		12:30	Suva Point to Flagstaff, Air conditioning on
	1 trip windows closed	12:30	Shopping centre, outdoor market stalls in carpark area.
Walk	Centrepont	14:45	Walking along Kings Wharf area near Suva markets
	City	16:15	Walking from the bus station in Suva City to the wharf
	Road City	16:00	Preschool, 100 m from road and elevated 10 m compared to road
	Kinoya	13:30	Walking mainly along a minor residential road and then major road (Kings Road).
	Road Kinoya	14:00	Lower campus of the University of the South Pacific (USP)
	Suva Point	10:00 to 12:30	Road from USP upper campus to USP lower campus
	Road Suva Point	09:30	4 walks in Flagstaff, near a busy intersection of roads
	Road Flagstaff	09:00	
		12:40	
		13:00	
		15:05	

October 2015 period were obtained for Nausori Airport (Weatherspark, 2016). Whilst Nausori airport is 17 km from the Suva City site, this was the nearest meteorological station for which cloud cover data were available. Pasquill (1961) developed a simple scheme to describe atmospheric stability, describing the tendency of an air parcel to move vertically; Class A, the most unstable, where vertical updrafts tend to develop; to class F, the most stable, where vertical updrafts tend to be suppressed. For Suva City, these stability classes were calculated according to the method of Gifford (1962) for the October 2015 period and averaged to give a typical daily occurrence profile (included as Supplementary Figure S1).

In order to compare these data to typical exposures in Suva, survey data were used. A survey of 125 Suva households was conducted in October 2014. This survey, intended to characterise emissions and exposure, is described in further detail in Isley et al. (2016). Survey results represent 25 different neighborhoods within Greater Suva, including 81 households from within the Suva City Council area. Demographic questions (including age, residential suburb and household size) were included to ensure a relevant population sample; questions and survey format were developed as part of the ethics approval for this project. Questions relevant to this study include mode of transportation used and commuting times via different modes of transport.

2.2. Instrumentation and quality control

2.2.1. Ultrafine particles

A Philips Aerasense Nanotracer was used to measure time-series of PNC and dp in the 10–300 nm size range. This size range exceeds the defined boundary of ultrafine particle range (diameter < 100 nm). Since the majority of the particles by number are typically within the ultrafine range, for simplicity, in this manuscript we refer to the Nanotracer data as ultrafine particles. Prior to sampling, the Nanotracer was calibrated in the laboratory using a condensation particle counter, as per the method of (Mazaheri et al., 2014). A PNC correction factor was derived for the Nanotracer at the start of the Suva project by running the PNC alongside a condensation particle counter measurements (TSI model 3787) in the laboratory at 16s intervals over 10 h. A second calibration test provided agreement with this correction factor (within 10%). Compared to condensation particle counter measurements (TSI model 3787) the Nanotracer may underestimate PNCs by up to 30% at high concentrations (above $1.2 \times 10^5 \text{ cm}^{-3}$),

but shows good agreement (within 10%) at PNCs below $8 \times 10^4 \text{ cm}^{-3}$ (Mazaheri et al., 2014). Since PNC observed in this study were mostly at levels less than 10^5 cm^{-3} , the potentially impaired performance the Nanotracer at high concentrations was not a significant concern.

2.2.2. Real-time PM

The Turnkey Osiris sampler was used to measure particle concentration of four size ranges of particles simultaneously, including PM₁, PM_{2.5}, PM₁₀ and TSP, recorded as 1-min averages. A heated inlet was used to precondition the particle sample and account for moisture derived artefacts; airflow was 0.6 L/min. The Osiris sampler uses a 25 mm stretched Teflon filter; this was weighed under controlled temperature (approx. 22 °C) and relative humidity conditions (approx. 50%) before and after sampling for the October 2015 period. As the Osiris records total mass collected between filter changes, this gravimetric mass was able to be used to calibrate the continuous PM measurements of the Osiris. Spinazzè et al. (2017) recommend use of a custom correction factor, as urban PM concentration measured by photometers may be up to five times greater than the gravimetric method. Gravimetric analysis of the Osiris filter provided a correction factor of CF = 0.29 (where gravimetric mass = photometer PM x CF). This Osiris had operated in Suva for 12 months; the correction factor varied by 6% during this time, based on monthly gravimetric analysis of Osiris filters (Isley et al., 2017b). The Suva City Osiris was co-located with a gravimetric PM_{2.5} sampler (discussed below), during 2014–2015 (Isley et al., 2017b). Average Osiris PM_{2.5} (corrected) showed reasonable agreement (typically within 12%) with gravimetric PM_{2.5} samples for days of concurrent operation, over one year. Osiris samplers were also used at Kinoya and Suva Point from October 2014 to October 2015 (Isley et al., 2017b).

2.2.3. PM_{2.5} and elemental concentrations

During the October 2015 period, gravimetric PM_{2.5} samples were collected daily at the Suva City site. Samples were also collected from October 2014 to October 2015 for 24-h periods (midnight to midnight) each Wednesday and Sunday. Stretched Teflon filters were used to collect samples, using an Aerosol Sampling Program (ASP) cyclone sampler, see Cohen (1996); operating at 22 L/min flowrate. The ASP samplers have shown to have good agreement with Beta Attenuation Monitors (Hibberd et al., 2013, 2016). Black carbon (BC) was determined at ANSTO

from these filters using the Laser Integrated Plate Method. Transmission measurements using a HeNe laser (wavelength 633 nm) were performed before and after exposure; assuming a mass absorption coefficient of $7 \text{ m}^2/\text{g}$ (Taha et al., 2007). The BC method has shown good agreement with the IMPROVE-A thermal optical method (Hibberd et al., 2016) and thermal desorption (Hibberd et al., 2013). Elemental concentrations were determined by ion beam analysis (IBA) and used for the characterisation of particulate sources (Cohen et al., 2014; Crawford et al., 2016; Manousakas et al., 2017). Four simultaneous techniques were applied on a 2 MV Tandetron accelerator: particle-induced X-ray emission (Al to Pb); particle-induced gamma-ray emission (Na, F, Al, Mg and Li); Rutherford backscattering (total C, N and O); and elastic recoil detection for (total H). One field blank was collected for every ten exposed samples. The average detection limit was $0.0036 \mu\text{g}/\text{m}^3$; ranging from $0.00025 \mu\text{g}/\text{m}^3$ for Ni to $0.043 \mu\text{g}/\text{m}^3$ for Na; with average error of 0.36% over all species. Results for October 2014 to October 2015 Suva samples are discussed in (Isley et al., 2017b) and (Isley et al., 2017a). Sources contributing to $\text{PM}_{2.5}$ in Suva were determined by positive matrix factorisation (PMF) using 2014–2015 elemental concentration data (Isley et al., 2017a).

Further gravimetric samples were collected from Kinoya (6 samples) and Suva Point (7 samples) during 2014–2015 using a portable Ecotech Microvol sampler. This sampler meets Australian air sampling standards AS/NZS 3580.9.9 2006 and AS/NZS 3580.9.10 2006. Polycarbonate filters (47 mm diameter) were exposed for approximately 24 h using an Ecotech Microvol, with $\text{PM}_{2.5}$ cyclone with flowrate of 3 L/min. Suva City Microvol data shows a statistically similar $\text{PM}_{2.5}$ concentration to concurrent ASP samples (Supplementary Table S1). Microvol $\text{PM}_{2.5}$ varied, on average, 6% from corrected Osiris $\text{PM}_{2.5}$ data (Supplementary Table S1) with the largest variation at Suva Point (see Graphical Abstract). These samples were subject to the same gravimetric and elemental analyses as the ASP samples, except that total H could not be analysed due to the use of polycarbonate filters. The lower flowrate used by Microvol samplers also resulted in proportionally higher uncertainty (compared to ASP samples) for calculated elemental aerosol concentrations.

2.3. Data analysis

Time series data for real-time instruments were collated into a single data-set. Synchronisation of instrument time-stamps was checked daily. In order to compare PNC and PM data, PNC data were converted to 1-min averages. Hourly averages were also created for comparison with Bureau of Meteorology wind data. Brief periods of data loss occurred due to the stopping of equipment to download data, as well as due to a 24-h interruption to the power supply; approximately six days of concurrent PM and PNC data were collected for the Suva City site.

For analysis of the mobile monitoring data, similar activities were grouped together for statistical analysis of these data, allowing different environments, such as walking or travel by bus, to be compared. Global positioning system location data were combined with PNC data allowing spatial distribution to be mapped, as well as provide a check to field notes of locations and activities.

3. Results

3.1. Ambient (fixed location) sampling

For the Suva City site, PNC, dp and PM concentration were averaged over the entire sampling period, with a summary provided in Table 2. Also included are gravimetric PM, black carbon

Table 2

Summary of PNC and PM concentration at Suva City. Mean values over the PNC sampling period in October 2015 as well as longer term data are shown.

Suva City	Study period		Long-term average ¹	
	All days	Sundays	All days ²	Sundays
PNC $\times 10^4 \text{ cm}^{-3}$	1.64 ± 0.02	1.2 ± 0.3	—	—
Dp (nm)	38.9 ± 0.1	37.9 ± 0.2	—	—
HVAS ³ TSP $\mu\text{g}/\text{m}^3$	—	—	41 ± 4^3	—
Osiris TSP $\mu\text{g}/\text{m}^3$	45.1 ± 0.4	42.1 ± 0.2	28.4 ± 0.2	27.4 ± 0.4
Osiris PM_{10} $\mu\text{g}/\text{m}^3$	21.3 ± 0.2	21.0 ± 0.7	15.2 ± 0.1	14.8 ± 0.2
Osiris $\text{PM}_{2.5}$ $\mu\text{g}/\text{m}^3$	11.6 ± 0.1	10 ± 1	11.8 ± 0.1	9.7 ± 0.1
Osiris PM_1 $\mu\text{g}/\text{m}^3$	4.0 ± 0.1	3.9 ± 0.4	3.00 ± 0.02	2.92 ± 0.04
ASP $\text{PM}_{2.5}$ $\mu\text{g}/\text{m}^3$	10 ± 1	6.6^4	8.6 ± 0.4	6.0 ± 0.3
ASP BC in $\text{PM}_{2.5}$ $\mu\text{g}/\text{m}^3$	2.7 ± 0.5	1.5^4	2.6 ± 0.2	1.7 ± 0.1

1. October 2014–October 2015 except for HVAS, which is from Garimella and Deo (2007).

2. ASP data is for all days except Sunday - largely Wednesdays.

3. High volume air sampling over a four-year period (Garimella and Deo, 2007).

4. Only one data point available.

and long-term PM concentration data for this site. Errors listed throughout this paper are the standard errors. Regarding relationships between the individual mass fractions, the PM_{10} fraction accounted for approximately 50% of TSP at Suva City. This is interesting and implies that there were a high proportion of large sized particles – dust – in the air of Suva. The $\text{PM}_{2.5}$ fraction was 54% of PM_{10} at Suva City; much of which (35%) was PM_1 . Considered together with BC, which accounted for 27% of gravimetric $\text{PM}_{2.5}$ concentration (Table 2), this indicates a significant contribution from combustion emissions. In comparison to previous data, $\text{PM}_{2.5}$ and BC concentrations were similar during the October 2015 period; TSP and PM_{10} concentrations were higher than 2014–2015, though in line with older data. All parameters show the pattern of lower concentration on Sunday (as per Isley et al. (2017b)).

3.1.1. Variation with time

Diurnal variation of hourly mean PNC, dp and PM fractions (TSP, PM_{10} , $\text{PM}_{2.5}$ and PM_1) averaged over 8 day at Suva City are presented in Fig. 1. Lowest hourly mean PNC was $1.07 \pm 0.09 \times 10^4 \text{ cm}^{-3}$, occurring at 13:00, also corresponding to lowest hourly $\text{PM}_{2.5}$ ($5.6 \pm 0.7 \mu\text{g m}^{-3}$). Peak hourly mean was observed at 18:00 for PNC ($2.33 \pm 0.07 \times 10^4 \text{ cm}^{-3}$) and 20:00 for $\text{PM}_{2.5}$ ($16.6 \pm 1.2 \mu\text{g m}^{-3}$). A small peak in PNC occurred at midday, corresponding to a drop in particle size. Day-to-day variation in the PNC is large, as seen in the 95% confidence interval (Fig. 1). Variation in PM concentration is greatest overnight, from 18:00 to 06:00. Despite this variation, morning and evening peaks are apparent in PNC. A similar trend is seen in the PM concentrations (TSP, PM_{10} , $\text{PM}_{2.5}$, PM_1), though TSP, PM_{10} and $\text{PM}_{2.5}$ remain high overnight, following the evening peak, decreasing at around 05:00. PM_1 concentrations decreased between 20:00 to 03:00. This diurnal PM variation is also seen in recent studies for Suva (Isley et al., 2017b).

3.1.2. Variation with wind direction

Wind and pollution roses, presented in Fig. 2, provide a summary of the particle characteristics that were measured for different wind directions at the Suva City site, averaged over the 8-day sampling period. During the sampling period, there were a high proportion of light winds ($<3 \text{ m s}^{-1}$) from the north through to northeast. Stronger winds ($>5.5 \text{ m s}^{-1}$) were mainly from the east-southeast. A very small percentage of winds were recorded from the west-southwest during the sampling period, with most winds being from the north clockwise through to southeast. At the Suva City site, ultrafine particles of a larger diameter ($>55 \text{ nm}$ hourly

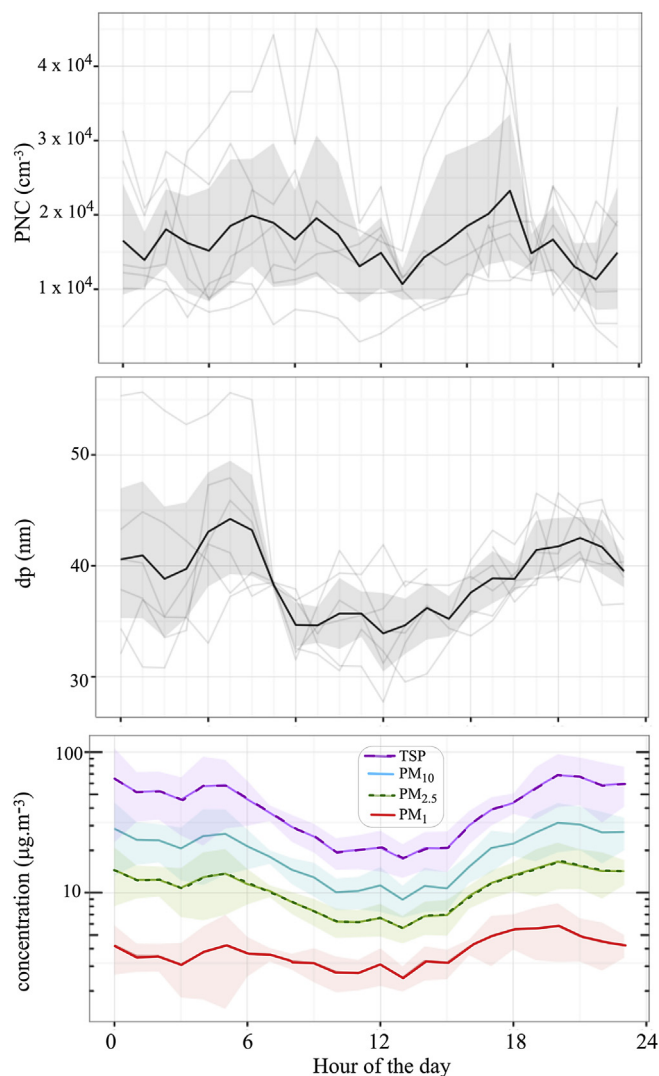


Fig. 1. Daily cycles of the mean PNC, dp and PM fractions (TSP, PM₁₀, PM_{2.5} and PM₁) at Suva City. Solid dark lines are the averaged data over 8 days and shaded regions show 95% confidence interval. Light grey lines for PNC and dp show the hourly average for each individual day.

average) were most frequently detected when winds were from the north. Ultrafine particles with $dp > 50$ nm were also detected during north-easterly winds. Smaller ultrafine particles (< 40 nm hourly average) were detected under a broad range of wind directions, from the north-west clockwise through to the southeast. Highest hourly average ultrafine particle concentrations of ($PNC > 10^5 \text{ cm}^{-3}$) were detected when winds were from the north-northeast as well as less frequent detections under north-north-westerly winds. PNC exceeding $9.0 \times 10^4 \text{ cm}^{-3}$ (hourly average) also occurred under winds from the east-southeast. During northerly and westerly winds, lower PNC ($PNC > 4.0 \times 10^4 \text{ cm}^{-3}$) occurred. For a large proportion of the time, hourly-averaged PM₁ levels remained below $6 \mu\text{g m}^{-3}$ for all wind directions. PM₁ concentrations were highest when winds were from the northeast, and for a lower proportion of the time, from the southeast and the east. PM_{2.5}, PM₁₀ and TSP pollution roses all appear to follow a similar pattern, with peak concentrations from the northeast and north-northeast. Concentrations under south-easterly conditions are generally lower, reaching about half of the north-easterly peak in each case.

3.1.3. Variation with wind speed

Particle concentration was also related to wind speed, however not as clearly as for wind direction. Whilst the p-value indicates a significant positive relationship between wind speed and PNC at Suva City ($p < 0.001$), the spread of data is very large (Supplementary Figure S2) and R^2 is very low ($R^2 = 0.12$). Hence, wind speed has only a very small influence in predicting PNC. Winds from the east-southeast, generally associated with lower concentrations of PM₁, PM_{2.5}, PM₁₀ and TSP, were typically stronger than other wind directions (Fig. 2), frequently exceeding 4 m s^{-1} . By contrast, winds from the north, northeast and north-northeast were characterised by much lower wind speeds, remaining below 4 m s^{-1} from the northeast and below 3 m s^{-1} from the north. Hourly profile data (Fig. 1) shows PM_{2.5} concentrations to be highest during the night-time hours. Night time wind speeds during the sampling period (Fig. 3) were between 1 m s^{-1} and 2 m s^{-1} ; whereas wind speeds during the day were generally above 2 m s^{-1} (Supplementary Figure S3). At the Suva City site, wind direction appears to play a larger role than wind speed in determining PNC. Plotting the night-time versus day-time winds as separate wind roses (Fig. 3), demonstrates this dependency. Night-time winds, as well as lower in speed, were generally from directions in the northeast sector. Daytime winds were predominantly from the south-southeast. The two parameters, wind speed and wind direction, must therefore be considered together.

3.1.4. Variation with atmospheric stability

The most prevalent stability class, experienced 36% of the time during the monitoring period, was class 'F', or stable conditions (Supplementary Figure S1). These class 'F' conditions occurred mostly between 18:00 and 05:00, meaning that dispersion of pollutants at night time was limited. The next most common was class 'C', or slightly unstable conditions, which occur 26% of the time; combined with 16% class 'B' conditions (moderately unstable), with 'B' and 'C' occurring mainly 06:00 to 16:00, meaning that much greater dispersion of pollutants would have occurred during the day. Mixing heights (Supplementary Figure S4) were consistently below 200 m at night (20:00 to 06:00) during the monitoring period. Mixing heights increased steadily after 07:00, reaching a maximum of approximately 1200 m in the early afternoon. A sharp decrease in mixing heights was then seen each evening from 19:00 to 20:00.

3.1.5. Sources of PM

Modelling for the October 2015 period shows sea aerosol (21%) and windblown soil (15%) to be the dominant PM_{2.5} sources (Fig. 4). Mass concentration (column chart) and percentage contribution (pie charts) of PM_{2.5} sources are shown for October 2015 period alongside those measured at the same site over one year. Detail on PMF analysis and source determination, is presented in Isley et al. (2017a). The PMF analysis used PM_{2.5} samples collected over 2014 and 2015, inclusive of the October 2015 period. PM concentrations attributable to secondary sulphates, smoke from open burning (waste burning) and fossil fuel smoke (diesel combustion by industry, shipping and for power supply) are consistent with longer-term results. Increased concentration of windblown soil and industry Ca (concrete and quarrying industries), road vehicles 2 and heavy fuel oil (large ships) have altered the percentage source contributions to total PM_{2.5} for the October 2015 period. The overall percentage contribution by combustion sources (road vehicles, open burning, heavy fuel oil and fossil fuel smoke) is lower for the October 2015 period (34% compared to 48% 2014–2015 (Isley et al., 2017a)).

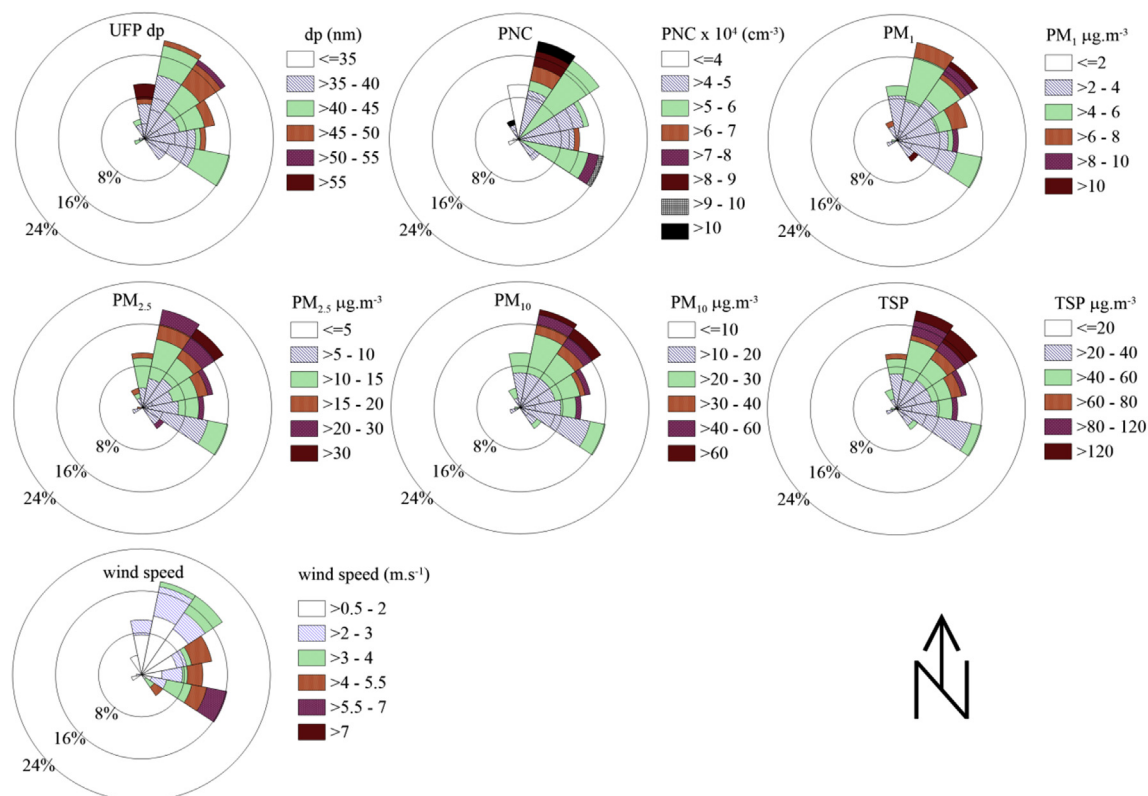


Fig. 2. Wind and pollution roses at Suva City site, showing frequency of counts of hourly-averaged data by wind direction during the 8-day sampling period in October 2015.

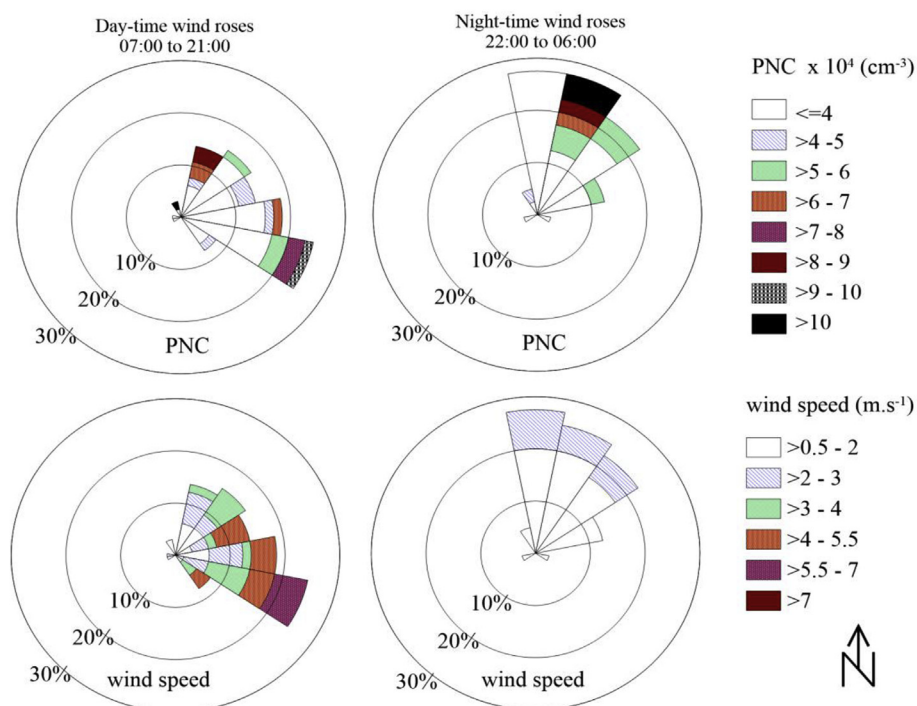


Fig. 3. Day-time and night-time wind roses and PNC pollution roses; showing frequency of counts of hourly-averaged data by wind direction during the 8-day sampling period in October 2015.

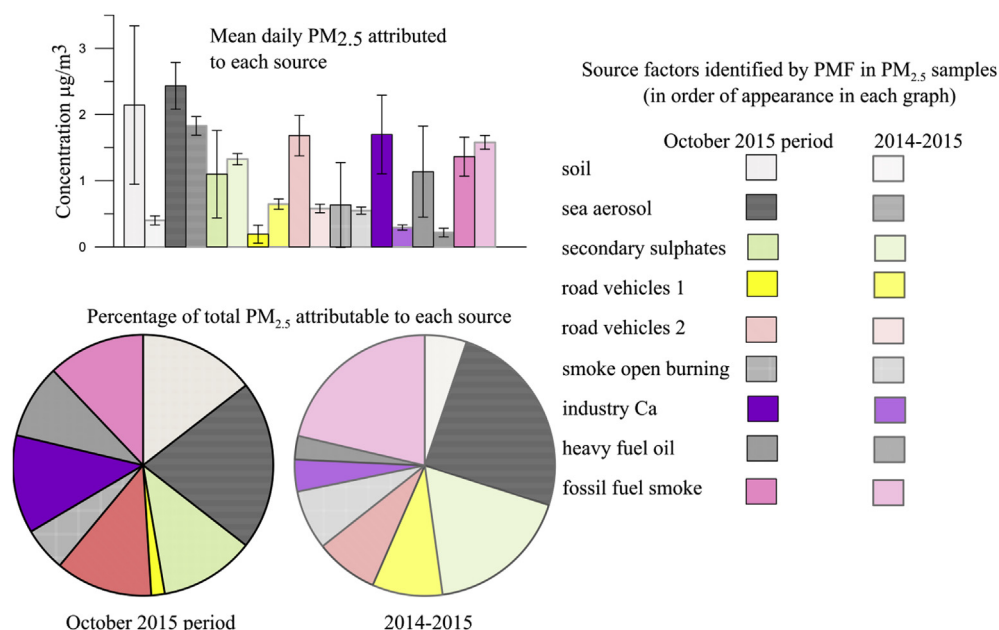


Fig. 4. Concentration and percentage contribution of PM_{2.5} sources at Suva City identified using positive matrix factorisation. Results for the October 2015 period are compared to those measured at the same site over one year. Data for 2014–2015 are from Isley et al. (2017a).

3.2. Mobile sampling

For mobile sampling, mean PNC ranged from $1.02 \pm 0.07 \times 10^4 \text{ cm}^{-3}$ whilst walking to $10.3 \pm 1.4 \times 10^4 \text{ cm}^{-3}$ whilst travelling on a bus. Greater detail regarding mobile sampling results is given in Fig. 5. Mean dp did not vary greatly (33 nm–39 nm) amongst the different microenvironments. For each location, boxplots show the median and range of PNC. Highest 16-s PNCs were recorded whilst travelling on buses ($73.4 \times 10^4 \text{ cm}^{-3}$), when walking ($51.4 \times 10^4 \text{ cm}^{-3}$) and travelling in taxis ($26.0 \times 10^4 \text{ cm}^{-3}$). Lowest concentrations were recorded whilst travelling in an air-conditioned taxi with the windows closed. The spatial variation of these data are presented on a map (Graphical Abstract) showing that highest levels were recorded at Flagstaff, Suva City and between Kinoya and Centrepoint. Lowest levels were recorded at Suva Point and whilst travelling by car between the airport and the city.

For Suva Point and Kinoya, additional data are available for comparison to PNC. Osiris PM, HVAS TSP, gravimetric TSP and PM_{2.5} and BC are included in Supplementary Table S1. At Suva Point, PM_{2.5} during the October 2015 period were higher than other data for this site. Data collected over 2014 to 2015 show Suva Point PM_{2.5} $5.5 \pm 0.8 \mu\text{g}/\text{m}^3$ (gravimetric) to be lower than Suva City, with BC ($0.2 \pm 0.2 \mu\text{g}/\text{m}^3$) at less than 10% of City levels (Section 3.1). Kinoya 2014–2015 gravimetric average PM_{2.5} ($12.7 \pm 1 \mu\text{g}/\text{m}^3$) returned higher values than Suva City, but with BC values at around half of Suva City levels (City $2.7 \pm 0.2 \mu\text{g}/\text{m}^3$, Kinoya $1.4 \pm 0.2 \mu\text{g}/\text{m}^3$).

3.3. Survey results

Survey results indicate that bus is the most popular mode of transport to work, as bus transportation is used by $67.7 \pm 6.0\%$ of people. Noting that people may use more than one mode of transport to work, $46.0 \pm 4.1\%$ of people travelled by car or taxi and $28.2 \pm 2.5\%$ by walking. Ignoring those who were unsure or did not answer the question, 71.8% of people indicated that they travel for some period of time each day by bus or car and 82.2% by walking or cycling. The typical time period spent each day travelling by car or

bus was approximately 30 min and approximately 26 min by walking.

4. Discussion

4.1. Ambient (fixed location) sampling

As previously mentioned, the perception of Suva and other Pacific Island cities is that, given the islands are small and isolated, with strong ocean winds and relatively low human populations, PNCs would be low. Mean ambient PNC, averaged over all 16 s readings for the October 2015 period for Suva City, $1.64 \pm 0.06 \times 10^4 \text{ cm}^{-3}$, is therefore higher than anticipated for Suva. A review of 24 urban PNC studies worldwide (Morawska et al., 2008), reports typical urban PNC of $1.08 \times 10^4 \text{ cm}^{-3}$, which is around 66% of the PNC measured in Suva. The Suva City PNC is similar to 'street' PNC levels in Basel Switzerland; where PNC (mean < 1000 nm) = $1.47 \pm 0.23 \times 10^4 \text{ cm}^{-3}$ (Ragettli et al., 2014) and urban areas in Taipei ($1.39 \pm 0.05 \times 10^4 \text{ cm}^{-3}$) Cheung et al. (2013); higher than ambient PNC averages reported for Basel (Ragettli et al., 2014), Prague, Vienna, Budapest (Borsós et al., 2012), Augsburg, Stockholm and Helsinki; but lower than Barcelona and Rome (Aalto et al., 2005). Whilst sampling time and spatial coverage of Suva were limited, these PNC results indicate that ultrafine particle concentrations in Suva are significance and require further consideration.

4.1.1. The role of wind speed in determining particulate dispersal

The daily PNC profile (Fig. 1) shows morning and afternoon peaks, although there is much day to day variation in these data. Peak PNC, at 18:00, corresponds with a wind speed decrease. Daytime wind speeds from 11:00 till 16:00 are approximately 4 m s^{-1} , decreasing to 3.3 m s^{-1} at 17:00 m s^{-1} and to below 2 m s^{-1} overnight (between 20:00 and 07:00). Hence peak-hour traffic conditions between 17:00 to 18:00, combined with a drop in wind speeds at this time, both contribute to this peak evening PNC. Atmospheric mixing height changes at 19:00 from around 1100 m to approximately 150 m, corresponding to mainly class 'F'

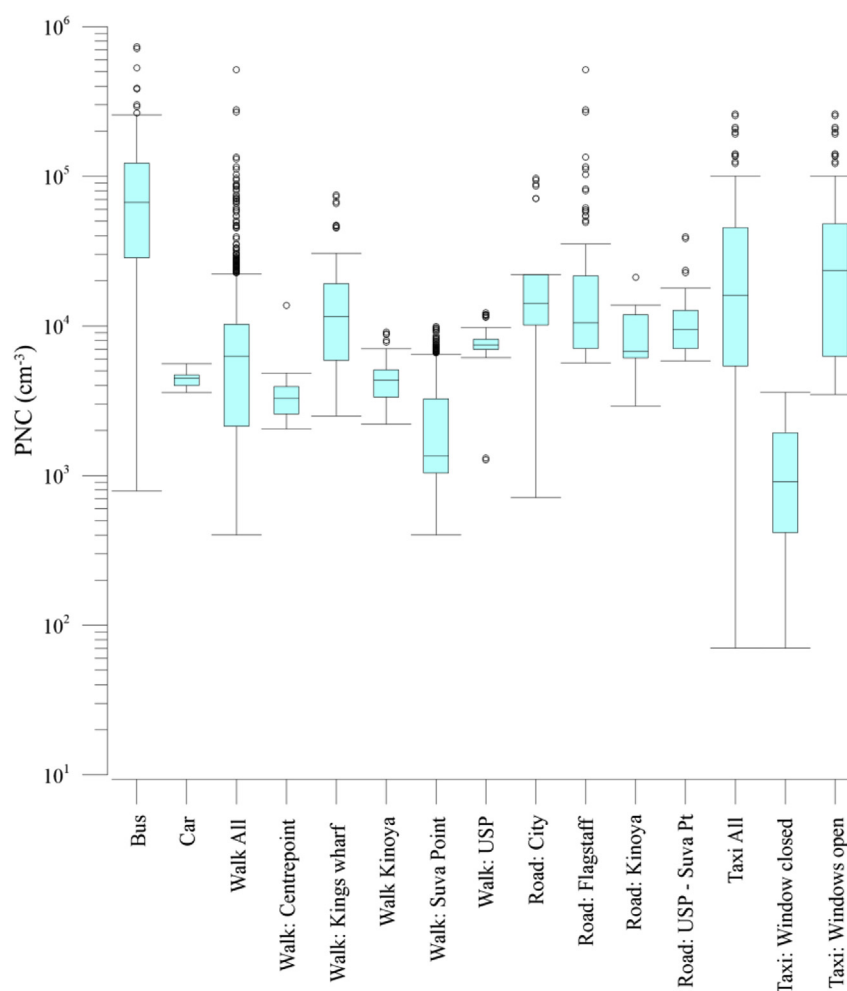


Fig. 5. Mobile sampling boxplots showing PNC for Suva microenvironments. Microenvironments labelled as 'road' denote walking alongside a road.

atmospheric stability throughout the night-time; impeding dispersion of pollutants overnight. This is seen in the elevated overnight concentrations of TSP, PM₁₀ and PM_{2.5}. These night-time dispersion conditions do not seem to be as significant for PNC, which shows a decrease after the evening traffic peak-hours (from 19:00), or similarly for PM₁, which decreases after 20:00. Whilst the morning PNC peak is less pronounced, maximum morning PNCs occur between 06:00 and 09:00, once again relating to peak vehicular traffic times (Fiji Roads Authority, 2015), indicating that vehicle exhaust may be a large component of ultrafine particle emissions in Suva. Wind speed does have some relationship with PNC and PM concentration, seen particularly in higher TSP, PM₁₀ and PM_{2.5} concentrations during low night-time wind-speed conditions. The relationship between wind-speed and PNC or PM at Suva City is less clear than the relationship with wind direction. Hourly concentration profiles (Fig. 1) and day/night wind roses (Fig. 3) show a pattern of lowest PM (TSP, PM₁₀, PM_{2.5} and PM₁) concentration in the middle of the day, which largely reflect the more favourable dispersion conditions at this time. Waste burning activities, throughout the evening in Suva's residential areas (Isley et al., 2016) also influence this diurnal cycle. Whilst PNCs decrease following the evening traffic peak, PNCs remain elevated overnight in comparison to midday levels (Fig. 1); with Suva displaying a similar daily PNC pattern to cities such as Vienna (Borsós et al., 2012), which have evening contributions from household heating.

4.1.2. The role of wind direction in determining particulate dispersal

Wind direction plays an important role in PNC and PM characteristics in Suva. Peak PNCs, as well as PM concentrations, occur when winds are from the built-up areas of Suva, combining traffic and industrial emissions with waste and biomass combustion activities. Highest hourly average PNC were detected when winds were from the north-northeast (Fig. 2), the direction of the Walu Bay industrial area and some of the busiest traffic areas of Suva (Graphical Abstract). Modelling of PM sources from elemental concentrations (Isley et al., 2017a) shows these wind directions to be associated with road vehicles, fossil fuel smoke and heavy fuel oil emissions. Steel industries at Walu Bay emit around 10 t yr⁻¹ PM_{2.5} from waste oil combustion (Isley et al., 2016); other Walu Bay industries; flour mills, beverage manufacture and other industries, would also contribute to PM and PNC, particularly from combustion of diesel. Further to the northeast (Graphical Abstract) are the two power stations at Kinoya, which burn diesel oil and heavy fuel oil; as well as a quarry and concrete manufacturing plant at Nasinu. Although much less frequent, hourly-averaged detections of PNC greater than 10⁵ cm⁻³ were also reported under north-north-westerly winds, potentially from shipping activities in Suva Harbour. PNCs >9.0 × 10⁴ cm⁻³ (hourly average) also occurred during winds from the city centre, an area characterised by large vehicular traffic volumes. PM₁ concentrations were highest when winds were from the northeast (Fig. 2), again being the direction industries (Walu Bay and Nasinu), heavily trafficked areas and

power stations. For a lower proportion of the time, PM₁ levels exceeded $8 \mu\text{g m}^{-3}$ from the east, the direction of the central Suva bus terminal and exceeded $10 \mu\text{g m}^{-3}$ from the southeast, the direction of city centre traffic. For PM_{2.5}, PM₁₀ and TSP, peak concentrations were, similarly, from the northeast and north-northeast. As well as exhaust emissions from traffic sources, the high traffic movements in these directions generate suspended road dust, particularly as many roads in Suva contain potholes and dirt edges and a number of roads and work-yards remain unpaved. The activities located in this direction are associated with burning of waste and biomass (Isley et al., 2017a), being areas where the residential population is concentrated. In comparison, the highest concentrations under south-easterly wind conditions, for PM_{2.5}, PM₁₀ and TSP alike, were only about half of those from the northeast.

4.1.3. Sources of particulates

Winds from all dominant wind directions primarily carried ultrafine particles in the 35 nm–50 nm size range (hourly averaged); hence a wide variety of traffic and industrial sources are likely to contribute to concentrations of these smaller diameter ultrafine particles. This relatively small particle diameter indicates that the PNC sampled in Suva represent fairly fresh ultrafine particles from local emission sources, rather than aged particles. Diesel is the most commonly used transport fuel in Suva, containing up to 500 ppm sulphur; and diesel vehicle emissions contribute around 100 t yr^{-1} PM_{2.5} to Suva's air (United Nations, 2012; Isley et al., 2016). A study by Ristovski et al. (2006) analysed emissions from diesel-fuelled buses of different ages, up to 19 years old; demonstrating that 58% of emitted particles were smaller than 50 nm for this fuel type (up to 500 ppm sulphur); indeed median particle diameters were typically in the 35 nm–50 nm size range. The mean dp for Suva City ($38.9 \pm 0.1 \text{ nm}$ mean over 8 days of 16 s readings) lies within this typical diesel exhaust size range. Ristovski et al. (2006) also reported that this higher sulphur content fuel had higher ultrafine particle emissions than the lower sulphur content fuel tested (50 ppm sulphur). It is therefore likely that diesel vehicle emissions in Suva contribute substantially to the PNC measured in this study. The central Suva bus terminal and busiest traffic areas of Suva are within 1 km of this Suva City site, although traffic emissions are common to all residential and industrial areas of Suva. Source apportionment studies (Fig. 4) have shown that fossil fuel smoke and road vehicle emissions are both significant contributors to PM in Suva. In a review of ultrafine particle emissions, Kumar et al. (2013) note that road vehicle sources typically comprise up to 90% of total ultrafine particles in urban environments. They also consider other ultrafine particle sources; listing road-tyre interaction, shipping emissions, aircraft emissions, domestic biomass burning, forest fires, waste incineration, power plants, cigarette smoking, cooking and photochemical nucleation amongst typical urban ultrafine particle sources. Many of these sources are significant for Suva, as evidenced in the modelling of PM_{2.5} sources (Fig. 4).

Modelling of PM_{2.5} for the October 2015 period (Fig. 4) shows contribution from vehicles, fossil fuel smoke (diesel), shipping and open burning; all sources which would contribute to PNC. On a mass basis, contributions from waste burning and diesel combustion sources during the October 2015 period were consistent with those during 2014–2015. The road vehicles 2 source includes suspended road dust, hence increased contribution is likely due to sampling during a period of dry weather combined with wind flow from highly trafficked areas (north-westerly winds). These rainfall and wind conditions also apply to the increased soil and industrial Ca contributions. Much of the increase in PM_{2.5} during the October 2015 period is due to soil and dust-based source factors, which are

unlikely to influence PNC (Friend et al., 2013; Brines et al., 2015; Posner and Pandis, 2015). Ships burning heavy fuel oil produce ultrafine particles (NABU, 2014; Ntziachristos et al., 2016). Thus increased contribution from heavy fuel oil combustion, during the October 2015 period, may mean that the calculated PNC levels overestimate the long-term average.

4.2. Mobile sampling

PNCs recorded whilst using the same mode of transport differ throughout Suva. The range of PNC recorded whilst walking outdoors (Fig. 5) is, for example, an order of magnitude lower at Suva Point (mean PNC = $0.25 \times 10^4 \text{ cm}^{-3}$) than those recorded in the city (mean PNC = $1.40 \times 10^4 \text{ cm}^{-3}$). Outdoor measurements were also strongly influenced by proximity to a road. PNC whilst walking near the road at Suva Point were, on average, 1.45 times higher than other outdoor locations at Suva Point. For the city and Kinoya, PNC near the road were 1.98 times higher than nearby outdoor locations. This is similar to the findings of Both et al. (2013) for Jakarta, Indonesia. Many of the highest readings occurred where there is a high volume of vehicular traffic, such as at Flagstaff, where peak PNC whilst travelling by bus were ($73.4 \times 10^4 \text{ particles.cm}^{-3}$); inside a taxi, with front windows open, peaked at $26.0 \times 10^4 \text{ particles.cm}^{-3}$; and whilst walking outdoors near the road ($51.4 \times 10^4 \text{ particles.cm}^{-3}$). These peak on-road and roadside mobile PNC for Suva approximately five times higher than similar peak PNC measurements made in Bhutan (Wangchuk et al., 2015), a developing country with similar population to Fiji. In contrast to this, PNC inside an air-conditioned car at Flagstaff peaked at a much lower $0.56 \times 10^4 \text{ particles.cm}^{-3}$. These results for Flagstaff would be indicative of other highly trafficked areas within Suva.

4.2.1. Variation of PNC in Suva

Whilst PNC conditions may vary over time, the short-term results presented here provide an indication of the range of ultrafine particle characteristics and sources for Suva. As identified in the data presented here in Sections 3.1 and 4.1, Isley et al. (2017b) observed that each day of the week shows a similar diurnal cycle of PM levels (TSP, PM₁₀, PM_{2.5}, PM₁). Mobile sampling occurred in daytime, non-peak hours (Table 1, excepting private car) when ambient PM concentration and PNC are lowest (Fig. 1 (Isley et al., 2017b)). Variation in traffic flow and differences between vehicles would impact on PNC results. It is anticipated that mobile sampling during peak hour may have yielded higher PNCs. Still, at any time in Suva, traffic conditions typically involve smoky vehicles; as most vehicles emit visible smoke (40% of cars and 90% of other vehicles (Campbell, 2004; Department of Environment Fiji, 2007)). Mobile sampling occurred on Monday and Tuesday, which are characteristic of PM concentrations for week days in Suva, with Sundays recording lower PM concentrations (Isley et al., 2017b). A weekday-Sunday difference was likewise observed for PM and PNC (Table 2). Relevantly, Sunday is a rest day in Fiji with reduced industry, commerce and traffic. Therefore it is anticipated that PNC would be lower if mobile sampling had occurred on a Sunday. Suva's PM_{2.5} sources also show this weekday-Sunday variation (Isley et al., 2017a). Sundays report reduced contributions from open burning, road vehicles, fossil fuel smoke, soil and industry Ca. Road vehicles, soil and industry Ca factors contribute more strongly during the dry season (April to October (Isley et al., 2017a)), as identified here (Section 4.1). Most of the combustion-based PM_{2.5}, except heavy fuel oil emissions (shipping) were, however, consistent during the October 2015 period with 2014–2015 Suva data. Elevated shipping emissions may cause some overestimation of PNC. Away from the port area, effects of shipping on mobile sampling results would be decreased. Therefore, the mobile sampling days provide a

representative snapshot of Suva under dry-season conditions, with further sampling required in wet seasons to confirm the uniformity of these results over the year.

Black carbon levels vary in a similar manner to PNC (Table 2 and Supplementary Table S1); with very low levels of both at Suva Point, moderate levels at Kinoya and higher levels at Suva City. Kinoya and Suva Point BC data are not concurrent with PNC. Still, they show, that the spatial distribution of PNC reported here fit a longer-term pattern of combustion aerosols in Suva.

Mobile sampling for differing modes of transport shows the impact of modes of transport on personal exposure, as the PNC results for various modes of transport differed substantially. It is of particular interest that the highest concentrations were recorded whilst travelling in buses. The majority of local transit buses in Suva do not have glass windows. Instead, windows are fully open, with loose-fitting plastic rain-shields that may be lowered during wet weather. Mean PNC whilst travelling in buses ($10.3 \pm 1.4 \times 10^4 \text{ cm}^{-3}$) are 3.7 times higher than roadside levels at Flagstaff and ten times higher than average outdoor levels across Suva. Bus passengers are exposed to emissions from road traffic, often emitted very close to where they sit in the bus, as well as to entrained emissions from the bus that they are travelling on. A recent study in Greece (Argyropoulos et al., 2016), showed on-road PNCs, measured on top of a mobile vehicle, to range from $0.81 \times 10^4 \text{ cm}^{-3}$ to $6.79 \times 10^4 \text{ cm}^{-3}$ across Thessaloniki; Suva's mean bus PNCs are 1.5 times larger than the highest of these on-road measurements. Similarly, Knibbs et al. (2011) reviewed 47 PNC exposure studies, worldwide, relating to different modes of transportation; Suva's mean PNC whilst travelling by bus was 2.5 times higher than bus exposure levels reported in the reviewed studies. Interestingly, exposure whilst walking in Suva was almost five times lower than the typical levels presented by Knibbs et al. (2011). Taxis are also a popular form of transport in Suva. Like Suva buses, many of these operate with windows open. Travel by taxi, namely those with open windows, recorded the second highest mean PNC of all activities in Suva, at $4.00 \pm 0.44 \times 10^4 \text{ cm}^{-3}$. The PNC difference between travel in taxis with windows open and closed (Fig. 5) is similar to that reported by Both et al. (2013) for car travel in Jakarta.

These high exposures whilst travelling by bus have significant implications in relation to health, with $67.7 \pm 6.0\%$ of Suva residents indicating that they commute by bus. Survey results indicated that the typical time period spent each day, by Suva residents, travelling by car or bus, was approximately 30 min and approximately 26 min by walking. Both et al. (2013) expressed concern for traffic officers that spend a greater proportion of their time near roads, hence increasing their exposure to ultrafine particle air pollution. This concern is relevant for Suva, where bus drivers and inspectors, roadside kiosk workers and traffic officers spend much of their time on or near busy roads. This would mean increased exposure to ultrafine particles.

5. Conclusion

This study has addressed the identified lack of data for ultrafine particle in the Pacific Islands through a case-study of Suva; providing a snapshot of PNCs in different settings and their relationship with PM. It has demonstrated that, although the islands are small and isolated, with strong ocean winds and relatively low human populations, PNCs in Suva; both ambient and in different mobile settings; are elevated to a level that warrant further investigation. Elevated PNC and PM concentrations were experienced when winds were from the heavily built-up areas of Suva where traffic emissions, industrial emissions and emissions from household burning of wastes and biomass would be the highest.

Analysis of PM_{2.5} sources identified road vehicles and combustion of diesel, wastes/biomass and heavy fuel oil to contribute significantly to Suva's particulate concentrations. Transport emissions are a major source of ultrafine particles in Suva, as demonstrated by the elevated PNCs recorded whilst travelling by bus and when walking beside roads; as well as by the daily PNC profile, which showed morning and evening peaks corresponding to peak traffic times. Investigation of PNC in wet season and of indoor PNC levels is also recommended. Suva is very similar to other Pacific Islands in terms of dependence on diesel, ageing vehicle fleet and prevalence of waste burning. These PNC data for Suva indicate that a reduction of emissions from combustion activities, particularly from vehicle exhaust, is likely to bring improvement of both air quality and health in Suva and likewise for similar Pacific Island environments.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2017.08.021>.

Ethics

Official approval to conduct this research in Fiji was granted on the 28th of April 2014, reference RA20/14 by Parmeshwar Mohan, Permanent Secretary for Education, National Heritage, Culture and Arts, Fiji and, on 12th March 2014, reference 12/2/1, by Eleni Tokadua, Ministry of Local Government, Urban Development, Housing and Environment, Fiji. Ethics approval was granted for this research by the Human Ethics Secretariat at Macquarie University (reference number 5201400557), dated 24th of June 2014.

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Chapter Three: Source contributions

Chapter Three presents the following five papers:

Paper Three: Isley C. F., Nelson P. F., Taylor M. P. 2016. Managing air quality in Suva, Fiji
European Journal of Sustainable Development, 5, 4: 242-254

Paper Four: Isley C. F., Nelson P. F., Taylor M.P. Atanacio A. J., Stelcer E., Cohen D. D.
Mani F. S. and Maata M. 2018. Reducing mortality risk by targeting specific air pollution
sources: Suva, Fiji. *Science of the Total Environment* 612C: 450-461.

Paper Five: Isley C. F., Nelson P. F., Taylor M. P., Williams A. A. and Jacobsen G. 2018
Radiocarbon determination of fossil and contemporary carbon contribution to aerosol in the
Pacific Islands. *Science of the Total Environment* 643: 183-192.

Paper Six: Isley C. F., Nelson P. F., Taylor M. P., Morrison, A. L. Atanacio A., Stelcer E. and
Cohen D. D. 2018. Ambient air quality and indoor exposure: PM_{2.5} implications for health in
Suva. *Air Quality and Climate Change* 52(1): 35-43.

Paper Seven: Isley C. F. and Taylor M. P. 2018. Air quality management in the Pacific
Islands: A review of past performance and implications for the future. *Environmental Science
and Policy* 84:26-33.

Following on from Chapter Two which quantified air particulate concentrations, this chapter focuses on particulate sources. Papers three to six focus on determining the contributions of various emissions activities to the total burden of fine particulate ($PM_{2.5}$) in Suva's air. Whilst these four papers each seek to answer the question of which activities are most significant in terms of particle air quality, the methods utilised to achieve this are diverse. Together they provide a comprehensive picture of particulate sources in Suva and their potential risk of harm to human health.

Paper three considers source contributions from an emission inventory perspective. The approach applied involves literature review and emissions calculation. Papers Four to Six start with air sampling and work backwards to determine emissions contributions. Techniques used include elemental concentration analysis, statistical modelling and radiocarbon analysis of the sampled particulates.

Emissions inventory data for black carbon was used to infer global warming potential (Paper Three). Quantitative source contributions were also used to calculate the health risk associated with each source (Paper Four). Further to Chapter Two, this Chapter explores the differential exposure across parts of the same city. In doing so, these data show that air quality, particle composition and hence health risk is variable across Suva.

Economic and other factors influence waste management, burning practices and vehicle emissions in PICs. These are discussed in Paper Seven, which provides recommendations for future air quality management in the Pacific Island region.

This chapter provides the most comprehensive assessment available of particulate sources in the Pacific Island region. Moreover, it demonstrates the significance of local emissions to the air quality and health of the region's inhabitants.

Paper Three

Managing air quality in Suva, Fiji

Isley C. F., Nelson P. F., Taylor M. P. 2016. *European Journal of Sustainable Development*, 5, 4, 242-254.

This paper introduces the exploration of particulate emission sources by looking at known information about different emissions activities. In order to understand the most significant sources, emissions from different activities need to be quantified. Quantification is also necessary if any reduction or increase in these emissions contributions is to be measured and understood.

This study shows that diesel emissions are the most significant contributor to both PM_{2.5} and black carbon. Emissions from the open burning of domestic wastes were also found to be a significant contributor to air quality in Suva. McDowall (2005) confirms the burning of waste in Suva, even where collection services exist, although this is at a lower (though unquantified) rate than in rural areas, where it is estimated that 70 % of household waste is burned. Based on waste generation and landfilling data (Government of Fiji 2014), 20% to 36% of household waste in Fiji's urban areas remains uncollected, instead being dumped or burned. This mirrors the situation across Pacific Island countries where:

'A significant amount of MSW generated in many [Pacific Island Countries] does not enter the municipal system, being illegally dumped on vacant areas or burned, resulting in significant environmental damage. In some countries, this amount may exceed 70% of the total [municipal solid waste] generated. This is a serious issue for many island countries' (Woodruf 2014).

Managing air quality in Suva, Fiji

C.F. Isley¹, P.F. Nelson¹ and M.P. Taylor¹

Abstract

Increased vehicular traffic and industry, added to widespread burning of wastes, contribute to Suva's fine aerosol particle air pollution (PM_{2.5}); particularly aerosol black carbon (BC). As well as potential improvement of health and amenity on a local scale, better management of air quality has implications for global atmospheric warming. Diesel vehicle emissions contribute 100 ± 30 t of PM_{2.5} to Suva's air annually, including up to 75 t BC. A survey of 125 Suva households, in 2014, showed that most Suva residents burn a portion of their household waste, contributing 50 ± 14 t PM_{2.5} per year, including between 2.1 t and 17 t BC; a combustion activity that is largely unnecessary. The estimated specific forcing potential due to Suva's black carbon emissions is between 50.5 TJ and 114 TJ annually. Emissions reductions may be achieved through various means, including alternative waste disposal practices and changes in fuel types and in fuel sulphur contents.

Key words: *combustion emissions, PM_{2.5}, black carbon, emission estimation*

1. Introduction

The population of Fiji is becoming increasingly urban. An estimated 53.7 % of Fiji's 889,000 total population (United States Government, 2016), live in urban areas, with approximately 25 % (218,000 people) living in the capital, Suva (Government of Fiji, 2012); making Suva the largest city in the tropical South Pacific Islands. Suva residents are primarily concerned about air pollution from vehicle exhaust, however they are also concerned about emissions from industry, open burning and incinerators (Government of Fiji, 2007; Isley, 2013). Lelieveld, Evans, Fnais, Glannadaki, and Pozzer (2015) report that premature mortality due to air pollution is around 50 % higher in urban environments, where population and emissions are most concentrated, than in rural areas. Whilst PM_{2.5} concentration in Suva typically remains within the World Health Organisation guideline of $10 \mu\text{g}/\text{m}^3$ (Isley et al., 2016; World Health Organisation, 2006); aerosol BC concentrations in Suva, at $2.2 \pm 0.1 \mu\text{g}/\text{m}^3$, are higher than the average levels reported for cities in Australia, New Zealand, England and Ireland and indeed are similar to those in larger, more industrialised cities in Germany (Isley et al., 2016). Considering the levels of BC in Suva, it is likely that combustion sources will account for a significant proportion of fine aerosol PM emissions. Oil combustion produces fine PM and diesel PM are typically less than $1 \mu\text{m}$ in diameter (Kittleson, F., & Johnson, 2004; Slezakova et al., 2013). In a study of six cities in the United States of America, Laden, Schwartz, Speizer, and Dockery (2006) found that combustion particles in PM_{2.5} from vehicles were associated with increased mortality, whereas fine particulates derived from

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crustal sources were not associated with increased risk. Although the toxicity of particles from different sources is yet to be defined, mortality estimates based only on total PM_{2.5} mass ‘*may underestimate the total effect of PM_{2.5} on mortality*’ p. 367 (Lelieveld et al., 2015). The International Agency for Research on Cancer (IARC, 2013) classified diesel engine exhaust as carcinogenic to humans (Group 1, see also California Air Resources Board, 2016), based on an association of diesel exhaust exposure with an increased risk for lung cancer. Black carbon also absorbs light, warming the atmosphere. Black carbon has an atmospheric lifespan of less than one year, meaning that climate impacts from emissions occur in the short-term (Bond, Zarzycki, Flanner, & Koch, 2011). The amount of energy added to the atmosphere by BC emissions (specific forcing potential - SFP) can be calculated, however, this does not relate directly to temperature change, as other factors such as the earth’s feedback response need to be considered (Bond et al., 2011). ‘*For Fiji’s urban areas, vehicle emissions are amongst the most common and offensive forms of pollution*’ (Government of Fiji, 2007), p. 10. Over 40 % of cars and 90 % of other vehicles in Suva were observed to emit visible smoke (Campbell, 2004). Vehicle emissions depend on factors such as vehicle age, maintenance and fuel type (Bluett, Dey, & Fisher, 2008; Zhang, Stedman, Bishop, Guenther, & Beaton, 1995). In this regard, it was acknowledged in Fiji’s submission to the United Nations Commission on Sustainable Development (Rogo, 2011) that second-hand vehicles of low grade and older diesel vehicles, which are common in Fiji, emit comparatively high levels of particulates. Rogo (2011) reported that emissions were exacerbated by the lower taxes on diesel fuel as compared with petrol in Fiji c.f. (Kittleson et al., 2004; Yasar, Rizwan, Bari, Kausar, & Khan, 2013). On a volumetric and calorific basis, diesel fuel imports for automotive use in Fiji are almost double those for unleaded petrol (Fiji Bureau of Statistics, 2014). In Fiji, diesel fuel must contain no more than 500 ppm sulphur (United Nations, 2012), compared to 10 ppm in Australia (Australian Government, 2015) and New Zealand (Mateparae, 2013); or 15ppm in the United States of America (United States Government, 2012). Penalties apply for excessively smoky vehicles in Fiji under the *Land Transport Act* (Parliament of the Fiji Islands, 2000) and there is some evidence of enforcement (High Court of Fiji, 2004). However, the United Nations Economic and Social Commission for Asia and the Pacific (United Nations, 2013), argues that these regulations are ineffective and ‘*virtually never*’ enforced. To encourage use of younger vehicles, with lower emissions, imports of cars, trucks and buses over four years old to Fiji were banned in 2008; although in 2009, this was downgraded to vehicles over eight years old (World Trade Organisation, 2014). Tariffs were also reduced on new buses from 27 % to 5 % and import excise from 15 % to 5 %, from 2009 until 2011 (World Trade Organisation, 2014).

In addition to vehicle emissions, diesel is a major fuel used by local industries and hence derived PM forms a significant component of Suva’s aerosols. Sixty-four percent of Fiji’s electricity is derived from hydroelectric sources and 1% from wind power (Fiji Electricity Authority, 2014). However, two thermal power plants (Figure 1) operate at Kinoya, in Suva city’s northeast. In 2010, these plants consumed a combination of 31 ML industrial diesel oil and 28 ML residual oil (Keruring van Elektrotechnische Materialen te Arnhem, 2012). The Fletcher Steel factory at Walu Bay in Suva’s west burns a further 3.07 ML of waste oil per year (McDowall, 2005). Ships visiting the Port of Suva also use diesel to

drive their primary and auxiliary engines. In 2010, the cost of diesel (including heavy fuel oil) and total fuel imports was equivalent to 9 % and 17 %, respectively, of Fiji's GDP (Fiji Bureau of Statistics, 2014). The reliance on oil for electricity generation is typical of Pacific Island countries (Dornan & Jotzo 2012; Pacific Energy Summit, 2013).

Due to the relatively small land area in Pacific Island countries, burning continues to be a convenient and practical method for waste disposal. Mataki (2011 p. 179) reported that in Honiara, the capital of the Solomon Islands, '*burning of wastes is a culturally entrenched behavior*' and that burning and burying of wastes seemed to be the only affordable and hygienic processes of disposal at the household level. Thaman (2003) similarly list smoke from fires for burning a diverse range of waste and open fires for cooking, along with tobacco smoke, to be the main sources of domestic air pollution in the Pacific Islands, including Fiji and that these emissions '*constitute a significant health hazard*' (Thaman, Morrison, & Thaman, 2003) p. 15. Burning of wastes is practiced both in urban and rural areas in Fiji, including Suva. Unregulated burning of wastes is also conducted by various industries and institutions in Suva.

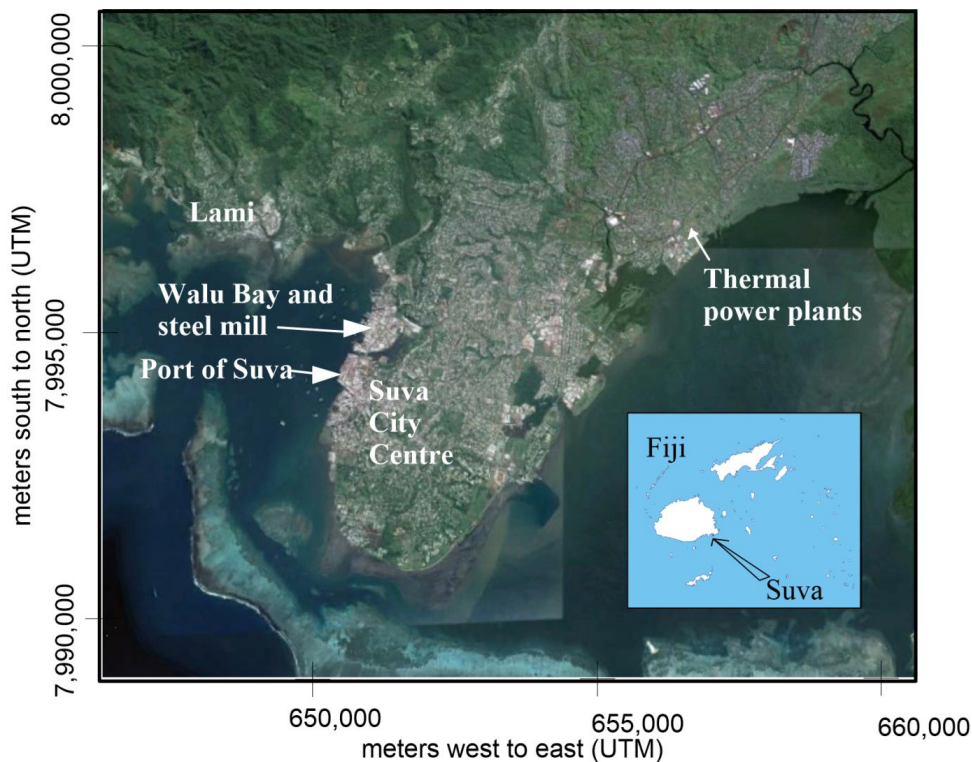


Figure 1: Location

2. Method

Household, industry and government combustion emissions were calculated

using survey data and government reports. In October 2014, 125 individual Suva households were surveyed to evaluate air pollutant emissions from household waste burning activities. Results represent 25 different neighborhoods within Greater Suva, including 81 households from within the Suva City Council area. Survey questions inquired as to whether council collected waste from the household, proportion of household rubbish and garden wastes burned and what materials were removed before burning. The literature sources used to compile the emissions inventory are detailed in Table 1. Unless pollution controls were documented, it has been assumed that emissions are uncontrolled. Incinerator conditions are, in some cases, outside of the scope considered in the emission rate documentation; one incinerator reports ‘*lit match*’ p.23 as the burner (McDowall, 2005), with paper as the fuel and another is fuelled by tyres, which are not included in reported combustion volume; hence emissions are likely to be under-reported. Emissions from fossil fuel combustion were based on import and usage statistics, as shown in Table 1. For waste oil emissions, no fuel quantity was explicitly stated in McDowall (2005), this has instead been inferred from boiler capacity, assuming continuous operation. The specific forcing potential (SFP) of BC emissions from Suva is directly related to BC emission; this has been calculated, for Suva, using (Bond et al., 2011)’s values for the Oceania region ($+1.03 \pm 0.56$ GJ/g BC).

Table 1: Data sources for fuel quantities and emission rates.

Emission source	Fuel quantity	Emission factor PM _{2.5}	Emission Factor BC as a proportion of PM _{2.5}
Household waste burning	Percentage burned (survey). Household waste quantity by province (Government of Fiji, 2012) Table 4. Waste composition (Government of Fiji, 2014) Table 3.	California Air Resources Board (2005); the same rate applies to both household waste (Table 2.5-1) and garden waste (Table 2.5-5), using an average of weeds, field crops, orchard and forest prunings.	4.2 % to 33 % (Chow, Watson, Lowenthal, Chena, & Motallebi, 2011) based on residential wood combustion.
Medical incinerators	Incinerator specifications for each hospital (McDowall, 2005)	Controlled air medical waste incinerators (United States Environmental Protection Agency, 1995a) Tables 2.3-2, 2.3-15	2.4 % (Chow et al., 2011)
Other incinerators	Specifications for quarantine, and government office incinerators (McDowall, 2005)	Controlled air solid waste incinerators (United States Environmental Protection Agency, 1995b) Table 2.1-9	2.4 % (Chow et al., 2011)
Industrial diesel and residual oil for electricity generation	Generator specifications by location (Keruring van Elektrotechnische Materialen te Arnhem, 2012)	National pollutant inventory (NPI) manual for boilers (Australian Government, 2011) Table 28 (industrial diesel), Table 26 (residual oil)	3 % to 13.6 % Chow et al. (2011), 77% (United States Environmental Protection Agency, 2012)

Waste oil combustion by industry	Furnace details for specific industries (McDowall, 2005; Secratiat of the Pacific Environment Programme, 2011); fuel conversion factors (United Kingdom Government, 2008). Assumed boiler operates continuously.	NPI boilers, (Australian Government, 2011) Table 30	77% (United States Environmental Protection Agency, 2012)
Industrial diesel imports not otherwise allocated	Fiji Bureau of Statistics (2014); assumed to be distributed according to the population density	NPI boilers (Australian Government, 2011) Table 28	77% (United States Environmental Protection Agency, 2012)
Combustion of fuel by vehicles	Fiji Bureau of Statistics (2012) Automotive diesel usage was assumed to be distributed according to vehicle registrations (Fiji Bureau of Statistics, 2012) and by population; light goods vehicles (LGV) 32 %, medium goods vehicle (MGV) 32 %, heavy goods vehicles (HGV) 31 % (assuming half of these are very HGV) and buses 5 %.	NPI combustion engines (Australian Government, 2008); using Table 10 unleaded petrol (ULP) and Table 14 for ULP with 10 % biofuel (same emission rate as ULP); diesel LGV (Table 15: 2.34 g/L), MGV (Table 20: 2.25 g/L), HGV (Table 21: 1.73 g/L), very HGV (Table 22: 1.12 g/L) and bus (Table 23: 1.12 g/L). These assume 10 ppm sulphur content, adjustment of + 32 % (Boulter & Latham, 2009) for Fijian vehicle fuels, which contain up to 500 ppm (United Nations, 2012)	Lower estimates listed are from Chow (2011): 5.9 % for ULP, 33% HGV diesel and 62 % LGV diesel; higher estimate of 75% is from USEPA (2012)
Liquefied petroleum gas (LPG)	Fiji Bureau of Statistics (2012)	Haneke (2001) Table 2	5.9 % to 37 % (Chow et al., 2011)
Kerosene burning	Fiji Bureau of Statistics (2012)	Based on wick stoves (Lam, Smith, Gauthier, & Bates, 2012), Table 5	80 % (Lam et al., 2012)
Household wood burning	Fuelwood consumption 0.05 m ³ per capita (Brown, 1997) Table 15, biomass density 0.57 t/m ³ (Food and Agriculture Organization, 2016); proportion of urban households cooking with biomass (survey).	California Air Resources Board (2005); as per garden waste above.	4.2 % to 33 % (Chow et al., 2011)

This study has focused on activities that occur within Suva and have potential for emissions reduction on a local scale. Emissions from shipping and aviation have not been included, although some shipping emissions may have been counted within industrial diesel calculations. Similarly, agricultural burning, which occurs away from Suva, has not been included. Emissions from property fires and cremation have not been calculated. Only primary PM emissions have been considered in this study; excluding secondary particles, formed in the atmosphere from emissions of SO₂ and NO₂. Where no error values were provided, European Environmental Agency (Pulles & Kuenen, 2009) methods for determining uncertainty in emissions calculations have been followed; these provide uncertainty calculations for comparing statistical data collected in different years as well as using emissions factors derived from various sources.

3. Results and Discussion

Survey results from October 2014 show that although 87.9 % of respondents have waste collection by council, more than half of Suva residents burn a portion of their household and garden wastes. The majority of households were able to estimate what proportion of waste they burn (Table 2), however 8.1 % of households were not sure of the proportion burned. Overall, 21.0 ± 3.8 % of Suva's household waste and 34.5 ± 6.2 % of garden wastes were burned. Suva council collected 109 kg waste per person in 2011 and estimated that 20 % of household waste remained uncollected (Government of Fiji, 2012). This waste stream is typically composed of 36.9 % kitchen waste with other major components being 21.4% garden waste (grass and wood), 15.3 % paper and 10.3 % plastic (Government of Fiji, 2014). The composition and the burning of wastes by households is typical of wastes across the Pacific Islands (Mataki, 2011), where variation of wastes across seasons and housing estates in differing socio-economic areas is not significant (Yao, 2008). Around half of households that burn waste remove metals and glass beforehand (Table 2). Considering these waste-stream proportions, the rate of waste burning in greater Suva is equivalent to each resident burning 27.7 ± 5.0 kg of household waste and 13.7 ± 2.4 kg of garden waste every year. This contributes 50 ± 14 t of PM_{2.5} into Suva's air every year, with between 2.1 t and 17 t of this being BC. Assuming that other areas with waste services exhibit similar burning behavior to Suva and using McDowall's (2005) rural waste estimates, household waste burning across Fiji contributes 210 ± 90 t PM_{2.5} annually, with up to 69 t BC.

Table 2: Summary of 2014 survey results for waste burning in Suva

Proportion burned (%)	Household waste (%)	Garden waste (%)	Waste material	Households removing material before burning as a subset of those that burn waste (%)
0	37.9	33.9	food scraps	32.3
< 50	37.9	13.7	plant materials	16.9
50	10.5	25.8	metals	50.0
> 50 to 100	1.6	5.6	glass	48.4
100	3.2	9.7	paper	8.9
not sure	8.1	8.1	plastic	37.1
Average	21.0	34.5	all waste is burned	2.4
			not sure	8.1

Considering that for Suva, most houses are serviced by waste collection, waste burning emissions could be greatly reduced. Only 21 % of respondents without waste collection indicated that they burn half or more of their household or garden waste. Alternative waste disposal methods practiced by communities not serviced by council waste collection in Suva include rental of skip bins on a community level, composting, feeding kitchen waste to animals, reuse of materials and local dumping of wastes. Metal and re-usable glass bottles can be exchanged for payment in Suva. The average proportion of waste burned by those without waste services is 45 ± 2 % of household waste and 43 ± 11 % of garden waste. Assuming that households in Suva without a waste service will require to burn 50 % of their waste, it is still possible to reduce the annual $\text{PM}_{2.5}$ emission from waste burning, in Suva, by 47 ± 12 t, including up to 16 t BC.

Using reported combustion volumes, incinerators contribute a combined annual total of 920 ± 230 kg $\text{PM}_{2.5}$ to Suva's air, including 22 kg BC. For Fiji, the annual emission is 2.3 ± 0.6 t $\text{PM}_{2.5}$, including 55 kg BC. The majority of incinerators are burning quarantine or medical wastes, hence finding an alternative hygienic disposal method is difficult. If alternate waste disposal were practiced for incinerators burning only paper (McDowall, 2005), an emission reduction of 93 ± 23 kg $\text{PM}_{2.5}$ per year could be achieved.

Households in Suva also burn kerosene or biomass fuel for cooking purposes. The October 2014 household survey reported that 28 % of Suva City Council households and 36 % of Greater Suva households surveyed used kerosene for cooking food. Many of these households use a combination of fuels, also including gas or electricity. The proportion of households burning kerosene for lighting were 17 % and 21 % respectively for Suva Council and the Greater Suva area. Kerosene represents an annual $\text{PM}_{2.5}$ emission of 420 ± 240 kg, with 33 ± 19 kg BC. While this is a small contribution to total $\text{PM}_{2.5}$ emissions, exposure is enhanced as indoor emissions are not readily dispersed. $\text{PM}_{2.5}$ emissions from LPG combustion are around half those for kerosene, at 170 ± 40 kg annually for Suva and 730 ± 180 kg for Fiji, with around one-tenth the BC emission of kerosene. Considering that 80 ± 7 % of Suva residents use LPG for cooking, this demonstrates that LPG provides a lower-emission alternative to kerosene. Likewise,

wood was used by 20 % of households, for cooking food, in Suva City Council and Greater Suva and coconut fiber was used by 5 %. Household wood burning for cooking contributes significantly to Suva's $PM_{2.5}$ with 13.4 ± 4.70 t $PM_{2.5}$ and up to 4.43 t BC emitted to Suva's air annually, with 203 ± 71 t $PM_{2.5}$ for all of Fiji. Contributions from diesel-fueled vehicles are far greater than for petrol vehicles (Table 3) and indeed are estimated to form the largest contribution to Suva's $PM_{2.5}$ and BC. On the whole, using fuel import volumes, vehicle emissions contribute 100 ± 30 t of $PM_{2.5}$ annually for Suva, including up to 75 t BC. For Fiji, this becomes 410 ± 100 t $PM_{2.5}$ including 310 t BC annually. Driving speeds, road gradients and vehicle maintenance all affect emissions (Grote, Williams, Preston, & Kemp, 2016); these factors are unable to be fully accounted for in these emissions calculations, which must therefore be considered as estimates. Boulter & Latham (2009) demonstrated that sulphur reduction from 307 ppm to 208 ppm (8.8 % sulphur reduction) in diesel fuel resulted in 10.5 % decrease in $PM_{2.5}$ emissions. For Suva, where sulphur may constitute up to 500 ppm (United Nations, 2012), it is expected that a 10 % decrease in sulphur content would reduce $PM_{2.5}$ emissions by approximately 10 %. This equates to a reduction of approximately 10 t per year $PM_{2.5}$, including up to 7.5 t BC for Suva. Reduction of sulphur content to 10 ppm is expected to give a 32 % emission reduction (32 t $PM_{2.5}$ annually based on Boulter & Latham 2009). Regulation of vehicle maintenance may bring additional emission reductions. Low sulphur fuels are more expensive and vehicle maintenance checks require personnel and equipment, forming an economic barrier to reduction of emissions. Further investigation is required into the cost of changing fuel composition and the comparative health costs from exposure to diesel combustion emissions.

Table 3: Emission from combustion activities in Suva and Fiji

Source	Annual quantity Suva ⁱ (Fiji)	$PM_{2.5}$ Emission rate ⁱⁱ	$PM_{2.5}$ emission t/yr Suva (Fiji)	BC emission Suva (t/yr)
Electricity generation industrial diesel (IDO)	31.0 ± 1.6 ML (58.2 ± 2.9 ML)	25.1 ± 5.0 kg/ML	0.78 ± 0.2 1.50 ± 0.4	0.024 to 0.60
Electricity generation residual oil	28.3 ± 1.4 ML (28.3 ± 1.4 ML)	0.108 ± 0.022 t/ML	3.1 ± 0.6 (3.2 ± 0.8)	0.092 to 2.4
Industrial waste oil furnaces	3.07 ± 0.15 ML (3.07 ± 0.15 ML)	3.26 ± 0.72 t/ML	10.0 ± 2.5 (10.0 ± 2.5)	7.7 ± 1.92
IDO industry other	39.7 ± 2.0 ML (162 ± 8 ML)	25.1 ± 5.0 kg/ML	1.0 ± 0.3 (4.1 ± 1.0)	0.80 ± 0.28
Incinerators medical waste	800 ± 40 t (1040 ± 50 t)	1.01 ± 0.20 kg/t	0.81 ± 0.2 (1.1 ± 0.3)	0.029 ± 0.001
Incinerators other	145 ± 7 t (1610 ± 80 t)	0.75 ± 0.15 kg/t	0.11 ± 0.03 (1.2 ± 0.3)	$0.0012 \pm$ 0.0004

Vehicles ULP	24.9 ± 1.3 ML (102 ± 5 ML)	62 ± 12 g/ML	0.0015 ± 0.004 (0.0063 ± 0.016)	$0.000091 \pm$ 0.000032
Vehicles diesel	$4.18 \pm 0.21 \times$ 10^7 L ($1.71 \pm 0.05 \times$ 10^8 L)	2.40 ± 0.48 g/L	100 ± 30 (410 ± 100)	40 to 75
LPG	4.37 ± 0.21 kt (18.2 ± 0.9 kt)	39.9 ± 8.0 g/t	0.17 ± 0.04 (0.73 ± 0.18)	0.0010 to 0.0064
Household waste burning	6270 ± 1200 t (26.2 ± 4.7 kt)	8.0 ± 1.2 kg/t	50 ± 14 210 ± 90	2.1 to 17
Household wood burning	1.68 ± 0.37 kt 25.3 ± 5.0 kt	8.0 ± 1.2 kg/t	13.4 ± 4.70 203 ± 71	0.56 to 4.43
Kerosene	1.00 ± 0.05 ML (4.00 ± 0.20 ML)	42 ± 24 g/L	0.42 ± 0.24 (0.17 ± 0.10)	0.33 ± 0.19
TOTAL			180 ± 53 845 ± 267	49 to 111

[i]. (Fiji Bureau of Statistics, 2012; Government of Fiji, 2012, 2014; Keruring van Elektrotechnische Materialen te Arnhem, 2012; McDowall, 2005; Pulles & Kuenen, 2009; Secretariat of the Pacific Environment Programme, 2011) [ii]. (Australian Government, 2008, 2011; Boulter & Latham, 2009; California Air Resources Board, 2005; Haneke, 2001; United Nations, 2012; United States Environmental Protection Agency, 1995a, 1995b). [iii] (Bond et al., 2004)

Industrial diesel emissions contribute a combined total of about 15 t of PM_{2.5} to Suva's air annually, including approximately 8.5 t BC. Waste oil emission rates assume a sulphur content between 0.01 % and 0.2 % (Australian Government, 2011); if sulphur contents are outside of this range, or emissions controls are present, the emission may vary from that reported here. A change from the use of residual oil to industrial diesel (IDO) would bring a reduction of around 2.4 t PM_{2.5} per year for Suva. Changing from use of waste oil to IDO could theoretically reduce emissions by around 9.9 t annually. Air quality is not the only consideration here. Combustion of waste oil by industry prevents disposal of these oils into waterways, where they can damage aquatic ecosystems and human health. If not combusted to provide benefit for industry, it is also possible, even likely, that waste oils would be combusted simply as a disposal technique. Likewise, the increased cost of IDO over residual oil, used for electricity generation, would increase the cost of production and ultimately the cost to electricity consumers. A variety of other emissions controls are available for industrial combustion, including pre-treatment of oils prior to combustion. The investigation of suitable emission control techniques and the economic, health and environmental consequences of using different fuels are necessary.

Diesel vehicles were calculated as the largest contributor to Suva's PM_{2.5} and BC, followed by household waste burning, household wood burning, industrial waste oil combustion and residual oil burning. Similarly, transport emissions and residential use of biofuels are amongst the largest contributors to BC in many areas of the world (Bond, 2007). Not all sources contribute equally to exposure and health impact. Industrial emissions are usually emitted via stacks, often at high temperature, where thermal buoyancy and dispersion of pollutants results in ground level concentrations that are

much lower than emitted concentrations. For household burning of wastes and vehicle emissions, emission points are much lower to the ground and nearby individuals are exposed to a greater proportion of the original emitted concentration. Emissions from cooking, which occur within the confines of a dwelling, have greater exposure potential. Unlike waste burning and vehicle emissions, industrial emissions tend to occur away from heavily populated residential areas, also reducing exposure. These spatial considerations do not apply to the specific forcing potential of the emissions, which remain constant.

The annual SFP of Suva's black carbon emissions is between 50.5 TJ and 114 TJ. Being directly proportional to BC emissions, this SFP is predominately due to diesel vehicle emissions. Suva's per-capita BC emission is 0.22 kg to 0.51 kg per year. This is less than or similar to per-capita emissions from South Asia, Western Africa or the Middle East and about half that of the USA, Oceania and Eastern Europe (Bond, 2007). This relatively low per-capita emission does not explain the relatively high BC concentrations measured in Suva and further investigation of BC contributions is required. Shipping and agricultural burning contributions require consideration as well as the formation of secondary PM particles. Other pollutants; carbon dioxide, methane, nitrous oxide and fluorinated gases, that contribute to climate change, have not been considered in this study. Being a nation composed of 106 inhabited islands, Fiji has incentive to reduce BC emissions, which contribute to global warming and change in sea levels.

Conclusion

The greatest reductions in PM_{2.5} and BC emissions would be gained from changes in diesel vehicle emissions. The second highest contributor is household burning of wastes. Apart from those without waste collection services, this is a preventable emission and could reduce PM_{2.5} in Suva's residential areas by 47 ± 8 t annually. The economic and health basis for reducing emissions warrants further exploration, along with further data collection to refine this emissions inventory.

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Paper Four

Reducing mortality risk by targeting specific air pollution sources: A case study on Suva

Isley C. F., Nelson P. F., Taylor M. P., Mani F. S., Maata M., Atanacio A. J., Stelcer E. and Cohen D. D. 2017.

Supplementary information in Appendix C

This paper provides a detailed study of particle composition, sources and health impacts of different PM_{2.5} components identified in Suva.

Relationship to emission inventory (Paper Three)

This study builds on the Paper Three, providing a different perspective on emissions sources by studying actual particle composition in Suva city. Source contributions determined from air sampling are often different to those determined by emissions inventory. Hibberd *et al.* (2013) offer a number of the reasons for this difference including atmospheric mixing and dilution of pollutants, distance from sources, deposition of particles as well as consideration of only primary aerosols in the inventory. In reality, sampled aerosols are often a mixture of primary and secondary particles.

Consideration of pollutant sources via air sampling also provides a check to emissions inventories. For example, Fujita *et al.* (1992) show that sampling studies may indicate underestimation of a pollutant source. This paper is in agreement with Paper Three that contends that fossil fuel (diesel) emissions are the major human-influenced PM_{2.5} source in Suva. However, this paper further illuminates the major sources of PM_{2.5} by showing that

diesel particulate emissions may not be primarily due to road vehicle transport, but include substantive contributions from industry, power generation and shipping.

The sources

Sources identified and their relative contribution to Suva's PM_{2.5} are summarised in

Figure 3.1. Nine PM_{2.5} sources were identified for Suva:

- sea aerosols (natural emissions from wind and wave activity)
- secondary sulphates (secondary particles formed from a mixture of ocean and human-influenced sources)
- smoke from fossil fuels (Figure 3.2 and Figure 3.3)
- two road vehicle sources (Figure 3.4)
- smoke from open burning (Figure 3.5)
- soil (Figure 3.6)
- an industrial signature high in calcium (Figure 3.7); and
- heavy fuel oil emissions (Figure 3.8).

Nine factor PMF model

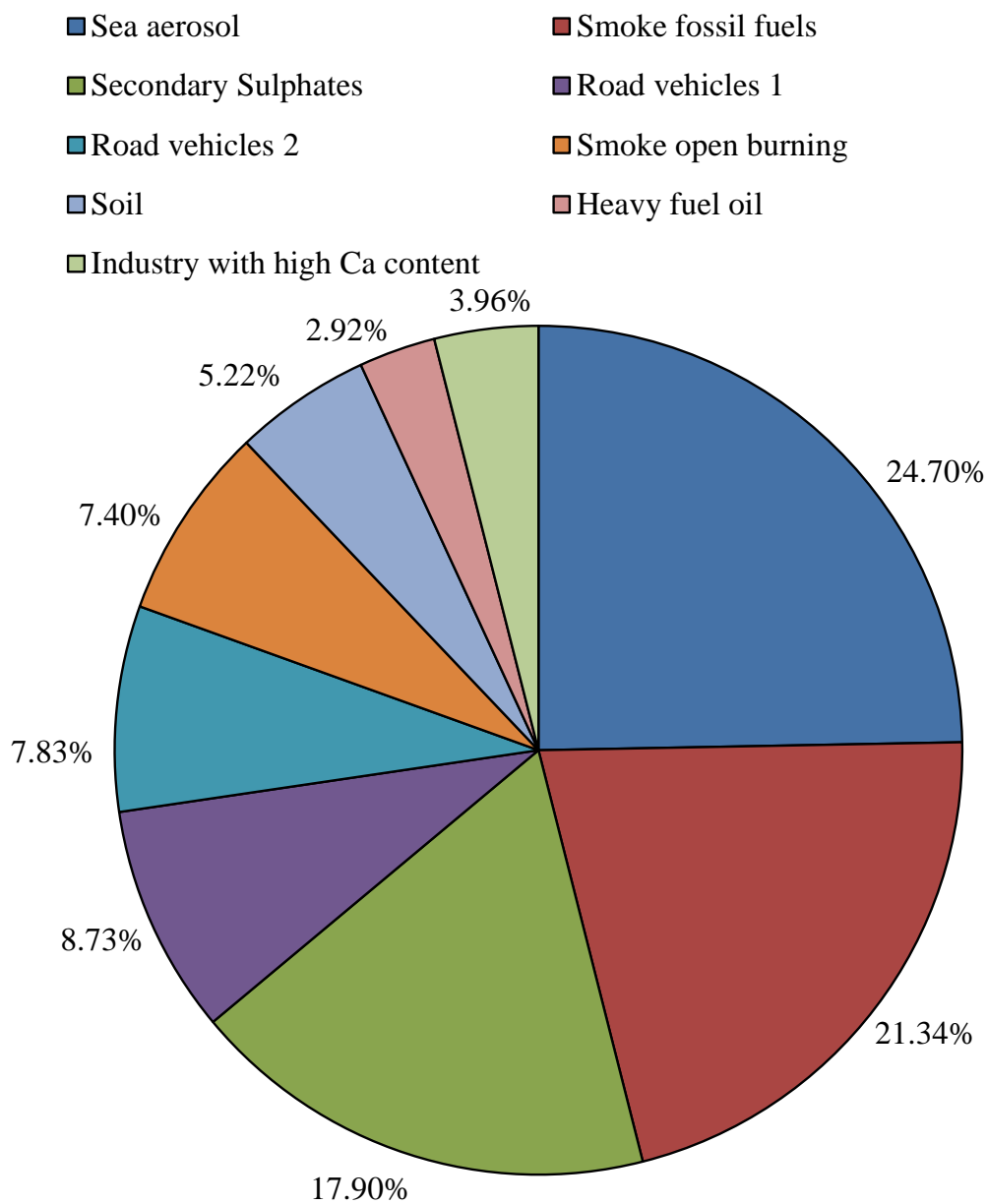


Figure 3.1 PM_{2.5} source factors identified for Suva city



Industrial activities at the Port of Suva (top) and Walu Bay (bottom and distant in top photo) burn diesel and waste oil. Other industries at Lami and Kinoya (including power generation) also contribute fossil fuel emissions (images dated April 2015)



Diesel generators are necessary due to power supply fluctuations and susceptibility to cyclones (Suva, November 2016). Two diesel-fuelled power plants also operate at Kinoya.

Figure 3.2 Smoke from fossil fuels - industry and generators



Small ships and motorised watercraft are a source of fossil fuel emissions (Kings wharf Suva, November 2016).



Even lawn-mowing (Fijians use brush cutters) contributes fossil fuel particulates (Suva, October 2015).

Figure 3.3 Smoke from fossil fuels – shipping and brush cutters



Figure 3.4 Road vehicles. With many used cars being imported and sold in Suva (left), congestion on Suva's roads (right) is increasing (images dated November 2016).



A waste pile (left) on the site of previous burning, ready to be burned, near Kinoya (October 2015). Right: Smoke from a waste fire drifts around houses near Lami (November 2014).



Left: A traditional lovo at Caubati (November 2014); meat and root vegetables are cooked slowly in the ground, surrounded by leaves. This method of cooking is not as common in the city as it is in villages. Right: Cooking with wood and coconut husk (April 2015) in a village west of Suva. This cooking method is also used in the Kinoya area.

Figure 3.5 Smoke from open burning. In Suva, smoke from open burning is likely to be largely from burning household waste and yard waste, but would also include smoke from traditional cooking practices.



Figure 3.6 Soil. Construction, vehicle movements on unsealed work yards, unsealed roads and roads with soft edges are a source of soil emissions (Suva, October 2015).

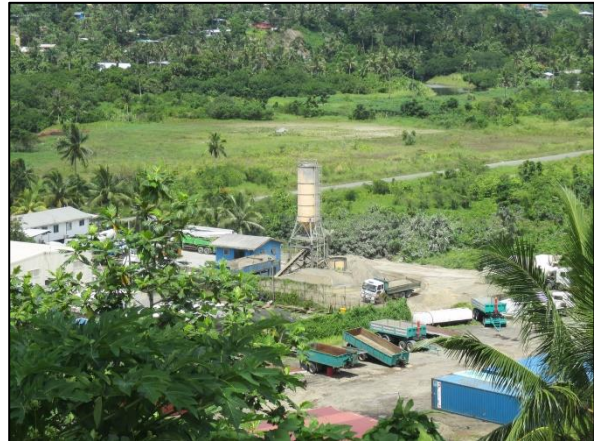


Figure 3.7 Industrial signature rich in calcium ‘Industry Ca’. The largest of Suva’s concrete plants (top and lower left), are based at Lami west of Suva. Smaller concrete plants, such as the one pictured lower right (also at Lami) are located throughout Suva. A quarry also operates at Nasinu (in Suva’s north). Images from Lami November 2016.

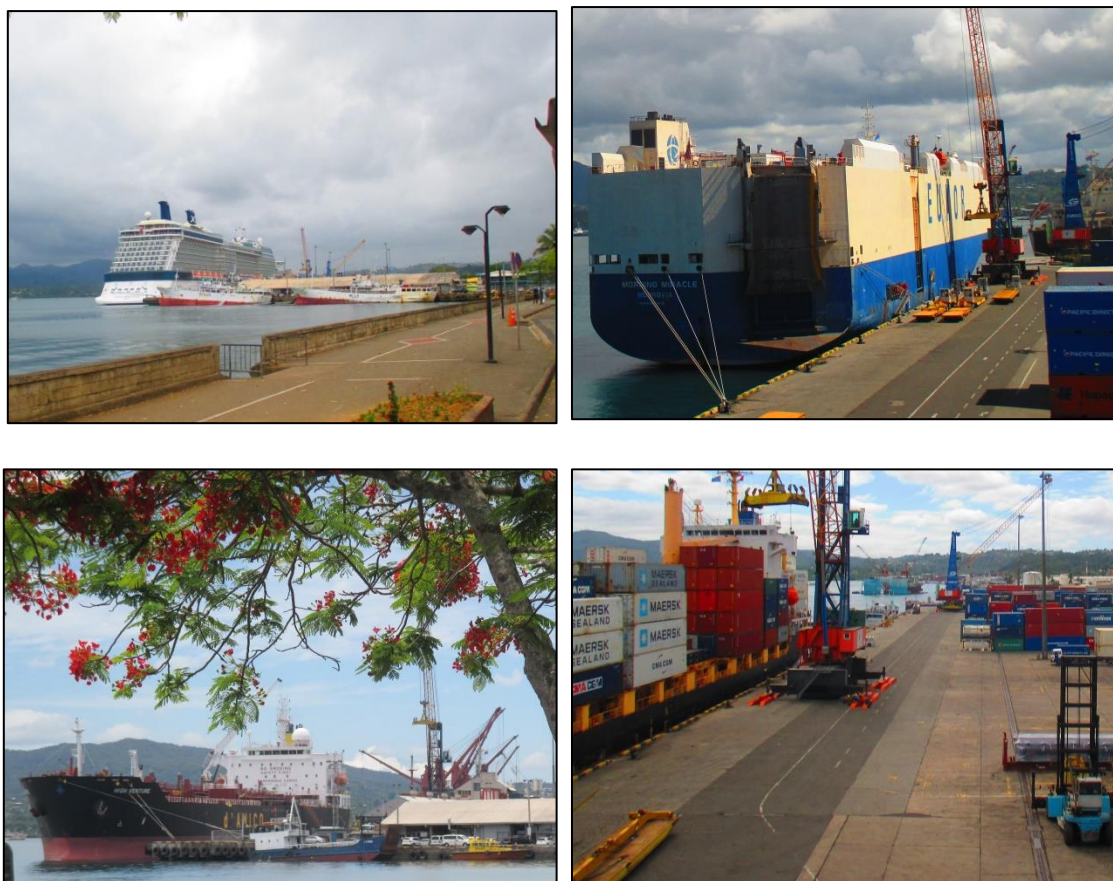


Figure 3.8 Heavy fuel oil. Heavy fuel oil emissions are from large ships. Suva is frequented by cruise ships and freight ships. Suva November 2016.

Health implications

The health effects of $PM_{2.5}$ are often insidious as Broome (2017) noted, ‘*no-one has “ $PM_{2.5}$ ” written on their death certificate*’. The damage caused by $PM_{2.5}$ is rather seen in its contribution to other health effects, which are detailed in Paper Four. Indeed, mortality is just one, albeit the most costly, potential consequence of $PM_{2.5}$ exposure. There are many lesser consequences that may go without notice, which can be described by the pyramid of effects (Figure 3.9; adapted from Larrieu *et al.* (2009). This paper does not quantify these lesser impacts for Suva, but focuses on the potential for reduced mortality that could be attained by reducing specific pollutant sources.

A summary of the available literature addressing the health impacts of PM_{2.5} components is provided in Table 2 of Paper Four. Relationship of PM_{2.5} sources to health impacts is a complex problem, due to the difference in PM_{2.5} mixtures from one location to another as well as findings that the toxicity of mixed PM_{2.5} components appears to differ greatly to that of individual components (Jia *et al.* 2017). Many studies cited in Paper Four relate to short-term exposures or specific demographic groups. These populations may experience environmental, dietary, healthcare access and economic factors that differentiate them from Suva, hence these studies were considered insufficient for making clear inferences about health risk (Wyzga and Rohr 2015).

The risk data for mortality implications of total PM_{2.5} concentration and black carbon content were considered more robust. Hoek *et al.* (2013) was used, as recommended by the World Health Organization (2013) and used in Broome *et al.* (2016). Hoek *et al.* (2013)'s risk estimates were based on a comprehensive global review of risk assessment studies. These are in also agreement with Chung *et al.* (2015)'s comprehensive epidemiological assessment regarding black carbon risk. Analysis of epidemiological associations between Fijian health statistics and air quality data are recommended to further understand the health risk of air pollutants in the local context.

For Suva, fossil fuel emissions contributions carry the highest risk to human health (Paper Four). Likewise, the Land Transit Authority Fiji (2015) noted that '*in any country smoke emission is seen as the silent road killer, at times surpassing the road accident toll.*' This paper compares air pollution health risk to Fiji's road toll and indicates that the impact of air pollution in Suva alone may indeed be higher than the national road toll in Fiji.

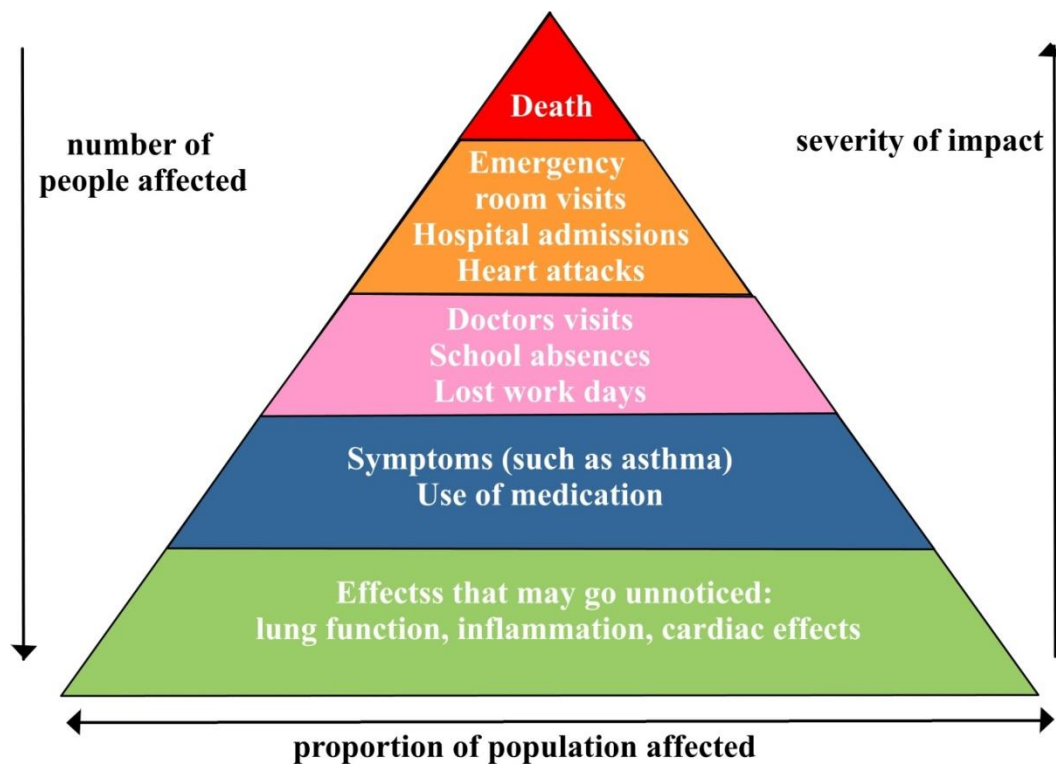


Figure 3.9 Health effects pyramid. Adapted from Larrieu *et al.* (2009).



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Reducing mortality risk by targeting specific air pollution sources: Suva, Fiji

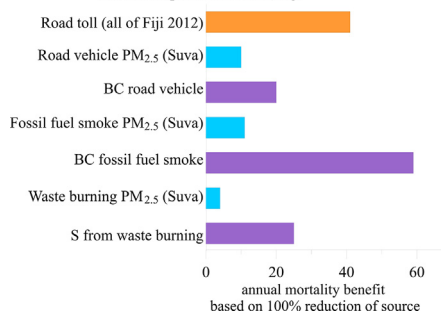
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HIGHLIGHTS

- Determines potential mortality reduction for different particulate matter emission sources
- Risk from black carbon and sulphur content exceed those based on particulate mass.
- Greatest benefits from reducing emission from fossil fuel, vehicles and waste burning

GRAPHICAL ABSTRACT

Predicted mortality benefit (ages >30 years) for reducing PM_{2.5} components in Suva, Fiji



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ABSTRACT

Health implications of air pollution vary dependent upon pollutant sources. This work determines the value, in terms of reduced mortality, of reducing ambient particulate matter (PM_{2.5}: effective aerodynamic diameter 2.5 µm or less) concentration due to different emission sources. Suva, a Pacific Island city with substantial input from combustion sources, is used as a case-study. Elemental concentration was determined, by ion beam analysis, for PM_{2.5} samples from Suva, spanning one year. Sources of PM_{2.5} have been quantified by positive matrix factorisation. A review of recent literature has been carried out to delineate the mortality risk associated with these sources. Risk factors have then been applied for Suva, to calculate the possible mortality reduction that may be achieved through reduction in pollutant levels. Higher risk ratios for black carbon and sulphur resulted in mortality predictions for PM_{2.5} from fossil fuel combustion, road vehicle emissions and waste burning that surpass predictions for these sources based on health risk of PM_{2.5} mass alone. Predicted mortality for Suva from fossil fuel smoke exceeds the national toll from road accidents in Fiji. The greatest benefit for Suva, in terms of reduced mortality, is likely to be accomplished by reducing emissions from fossil fuel combustion (diesel), vehicles and waste burning.

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1. Introduction

It is well established that increased $PM_{2.5}$ (particulate matter with equivalent aerodynamic diameter 2.5 μm or less) concentrations relate to adverse health impacts. Short and long term exposures to even low levels of $PM_{2.5}$ have been associated with all-cause mortality (Kloog et al., 2013; Shi et al., 2016), with no evidence of a threshold below which effects are not observed (Pope and Dockery, 2006; Brook et al., 2010). Hoek et al. (2013) reviewed available studies, finding that the risk of premature mortality due to $PM_{2.5}$ increases by 6% per $10 \mu g/m^3$ increase in $PM_{2.5}$; 11% for cardiovascular mortality. As coronary heart disease is the leading cause of death in Fiji (Ministry of Health and Medical Services, 2015), health implications of air quality warrant investigation. Associations have been made between $PM_{2.5}$ and diabetes (Potera, 2014; Rao et al., 2015), also a major cause of death in Fiji (Ministry of Health and Medical Services, 2015); deep vein thrombosis and pulmonary embolism (Kloog et al., 2015), as well as dementia, Alzheimer's and Parkinson's (Kioumourtzoglou et al., 2016). Health care access and health education also influence susceptibility to impacts arising from $PM_{2.5}$ exposure (Garcia et al., 2016).

Certain $PM_{2.5}$ constituents are more harmful than others (Bell et al., 2014; Dai et al., 2014; Chung et al., 2015; Kioumourtzoglou et al., 2015; Jia et al., 2017). In a toxicology study, Roper et al. (2017) reported the most significant associations of $PM_{2.5}$ with lung tissue inflammatory response from sampling sites with the lowest $PM_{2.5}$ concentrations; demonstrating that $PM_{2.5}$ concentration alone is not a satisfactory measure of health risk (similarly, Hao et al., 2016). Understanding of the health risk of $PM_{2.5}$ components, and the mechanism for toxicity, is still developing (Huang et al., 2014; Atkinson et al., 2015; Gray et al., 2015; Tong et al., 2015; Weber et al., 2015). Recent research has explored the role of oxidative potential, relating it to cardiovascular and respiratory health impacts (Weber et al., 2015; Lakey et al., 2016; Pei et al., 2016; Sarnat et al., 2016; Weichenthal et al., 2016; Yang et al., 2016). Weber et al. (2015) reported vehicle emissions and biomass burning sources to exhibit higher oxidative potential and hence higher toxicity than other sources. Air pollution control measures should include consideration of components, particularly those with high toxicities, rather than $PM_{2.5}$ mass alone (Liu, 2016). In particular, Eklund et al. (2014) advocate use of black carbon (BC) in air quality regulations, as an indicator of adverse health effects, due to the consistency of observed associations.

The disparate toxicity of $PM_{2.5}$ components is of relevance for the Pacific Islands, where diesel combustion and waste burning contribute significantly to the particulate loading (Periathamby et al., 2009; Mataka, 2011; Owens et al., 2011; Wiedinmyer et al., 2014; Escoffier et al., 2016; Taibi et al., 2016). For Suva, $PM_{2.5}$ levels are within World Health Organization guidelines ($7.4 \pm 0.3 \mu g/m^3$ annual average of Wednesdays and Sundays) yet black carbon in $PM_{2.5}$ ($2.2 \pm 0.1 \mu g/m^3$) is similar to levels in much larger cities (Isley et al., 2017) highlighting the need for further investigation of air pollutant sources in order to facilitate more effective air quality management. Source contributions to $PM_{2.5}$ for Suva would be typical of dominant of emission contributions across all the Pacific Islands. Similarly, other developing communities in Africa, Asia and South America that share similar climate, burning practices and usage of diesel are likely to be equally affected.

An emissions inventory (Isley et al., 2016) showed diesel vehicle emissions to be the largest source of BC in Suva. Emissions data was difficult to obtain, meaning that sources may have been underestimated. Calculated per-capita BC emissions for Suva were comparable to regions with much lower ambient BC than Suva, indicating that the inventory may not fully represent local emissions (Isley et al., 2016). Further data are required to accurately define emission contributions. Alternatively, elemental concentration data may be used to statistically model sources. This has been attempted by Garimella and Deo (2007); who interpreted elemental concentration data in Suva to provide qualitative conclusions about PM sources. By comparing PM concentrations to the earth's crust, Garimella and Deo (2007) concluded that Suva's PM was

dominated by marine aerosols, with possible inputs from automobile exhaust. Source apportionment in Garimella and Deo's (2007) study was constrained by identification of only 5% of total PM mass. This study aims to further that work, using a suite of elements likely to identify a larger proportion of particulate mass; H, Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, Sr, Pb, N and BC. Because of the well-defined health implications identified above, this study focuses on $PM_{2.5}$. In order to make the study data more specifically applicable to future Fijian air policy and health risk assessments, $PM_{2.5}$ sources will be defined quantitatively using the positive matrix factorisation (PMF) method.

The PMF technique (Paatero and Tapper, 1994; Paatero, 2000b; Paatero, 2000a) identifies statistical groupings of elements (source factors) within total $PM_{2.5}$ mass; which are then related to emission sources using known elemental ratios. Many recent source apportionment studies have used PMF (Belis et al., 2013; Karagulian et al., 2015), particularly due to its differentiation of sources that share common elements. This is an important advantage for Suva, where many combustion sources share BC, H and S.

The objective of this paper is to quantitatively identify source contributions to the total $PM_{2.5}$ burden of Suva's air and to determine the mortality risk presented by these factors. This will enable more effective and targeted air quality policy in Fiji.

2. Method

2.1. Sampling

Sampling was carried out in Suva city centre, at $-18.13^\circ S$, $178.42^\circ E$. On the west coast of the Suva peninsula, the site is located atop a 4-level building, at approximately 18 m height and 20 m elevation. The city markets, Suva bus terminal, Walu Bay industrial area and shipping port activities all lie within 1 km of this site. This site was selected as it is downwind of Suva City and would indicate the sources that people living and working in Suva are exposed to. Meteorological data (Australian Government, 2016) were obtained from instruments co-

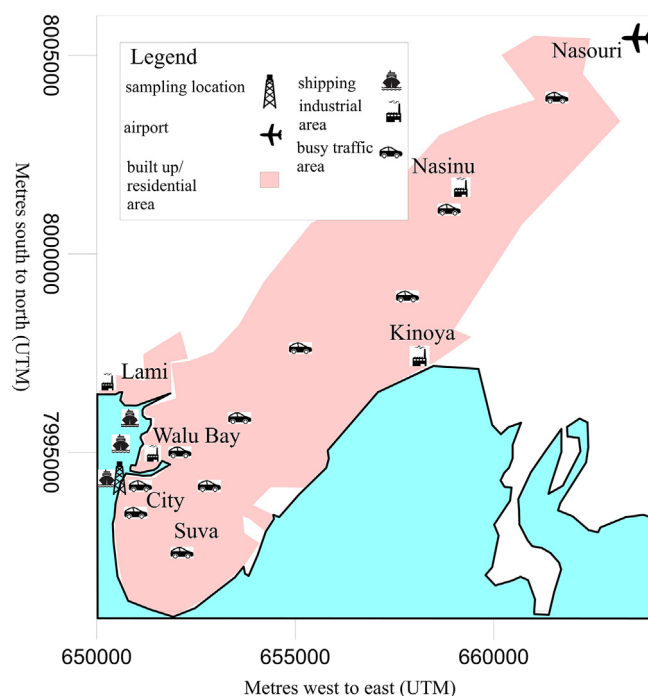


Fig. 1. Sampling locations, showing the $PM_{2.5}$ sampling location in Suva city centre as well as locations of major industries and other emission sources.

located at the site. A map of the sampling site, showing significant local sources is shown in Fig. 1.

Samples were collected for 24-hour periods (midnight to midnight) between November 2014 and October 2015 each Wednesday and Sunday. In addition, intensive campaigns covering approximately three weeks in total, allowed for more frequent sample collection during April/May 2015 and October 2015. Stretched Teflon filters (15 mm) were used to collect samples, using an Aerosol Sampling Program (ASP) cyclone sampler, built by the Australian Nuclear Science and Technology Organisation (ANSTO) according to the IMPROVE design (Cohen, 1996) and operating at 22 L/min flowrate. The ASP samplers have been shown to have good agreement with Beta Attenuation Monitors (BAMs) (Hibberd et al., 2013; Hibberd et al., 2016). One field blank for the ASP PM_{2.5} sampler was collected for every ten exposed samples.

2.2. Laboratory analyses

Concentration of PM_{2.5} was determined by weighing filters under controlled temperature (approx. 22 °C) and relative humidity conditions (approx. 50%) before and after exposure. Black carbon (BC) was determined from the filters using the Laser Integrated Plate Method (LIPM). The BC concentration is estimated from two transmission measurements from a HeNe laser (wavelength 633 nm) performed on each filter before and after exposure; assuming a mass absorption coefficient value of 7 m²/g for carbon particles (Taha et al., 2007). This BC determination method, performed at ANSTO, has shown good agreement with IMPROVE-A thermal optical methods (Hibberd et al., 2016) and thermal desorption methods (Hibberd et al., 2013). Isley et al. (2017) provide a thorough analysis of gravimetric PM_{2.5} and black carbon results for Suva (the same data used in this study), including detail on quality control and comparison to other measurement methods.

Elemental concentrations were determined by ion beam analysis (IBA), a non-destructive technique established for the characterisation of particulate sources (Crawford et al., 2016; Manousakas et al., 2017). Analysis was carried out at ANSTO on a 2 MV Tandemron accelerator. Four simultaneous techniques were applied: particle-induced X-ray emission for concentrations of elements from Al to Pb; particle-induced gamma-ray emission for Na, F, Al, Mg and Li; Rutherford back-scattering to determine total C, N and O; and elastic recoil detection for total H. The application of these methods to air pollution source apportionment are discussed in Cohen et al. (2014); including the use of the PIXAN analysis code, the technique used here to extract the elemental concentrations from the spectrum peaks. Examples of spectrum graphs for Suva samples and blanks as well as calibration details and plots for ion beam analysis are included as Supplementary Section C. Average field blank levels of the analysed species in the particle samples were equivalent to 0.0015 µg/m³ (<0.02 µg/cm² on filters) with the majority of field blank values were below the method detection limit. The average detection limit for the particulate matter species was 0.0036 µg/m³; ranging from 0.00025 µg/m³ for Ni to 0.043 µg/m³ for Na; with average error of 0.36% over all species. Error and method detection limit matrices were inputted to the PMF analysis.

2.3. Determination of source factors

Positive matrix factorisation (PMF) was conducted at ANSTO, using the original DOS version of the PMF analysis codes (PMF2-DOS) developed by Paatero and Tapper (1994). These have been demonstrated to show good agreement with other PMF versions (Ramadan et al., 2003; Kim and Hopke, 2007; Kim et al., 2010; Hibberd et al., 2013). PMF utilises a measurement matrix $X(n,m)$ [n samples, m chemical species] which is factorised into smaller matrices $factor(p,m)$ [p factors, m elements] and $contribution(n,p)$ [p factors for each of the n samples]; each constrained to positive values. Then $X(n,m) = factor(p,m) \times contribution(n,p) + error(n,m)$; with the error matrix minimised in the PMF process. The PMF analysis was performed multiple times, each

time incrementing the number of source factors. The criteria used to determine a suitable fit for the source factor analysis were: all average factor contributions to total PM_{2.5} mass were > 1%; all factors were positive; all regression coefficients were positive; all p -values were < 0.05 and χ^2 is close to one. These statistical criteria were satisfied when eight source factors ($\chi^2 = 0.997$) or nine source factors ($\chi^2 = 0.748$) were assumed.

The eight and nine factor PMF model scenarios provided similar results. The nine-factor model is discussed in this paper. Elemental contributions to each factor in the eight factor model are included, for comparison, as Supplementary Fig. A1; further comparison between eight and nine factor scenarios is provided throughout the supplementary material. Uncertainty due to differences in modelled source

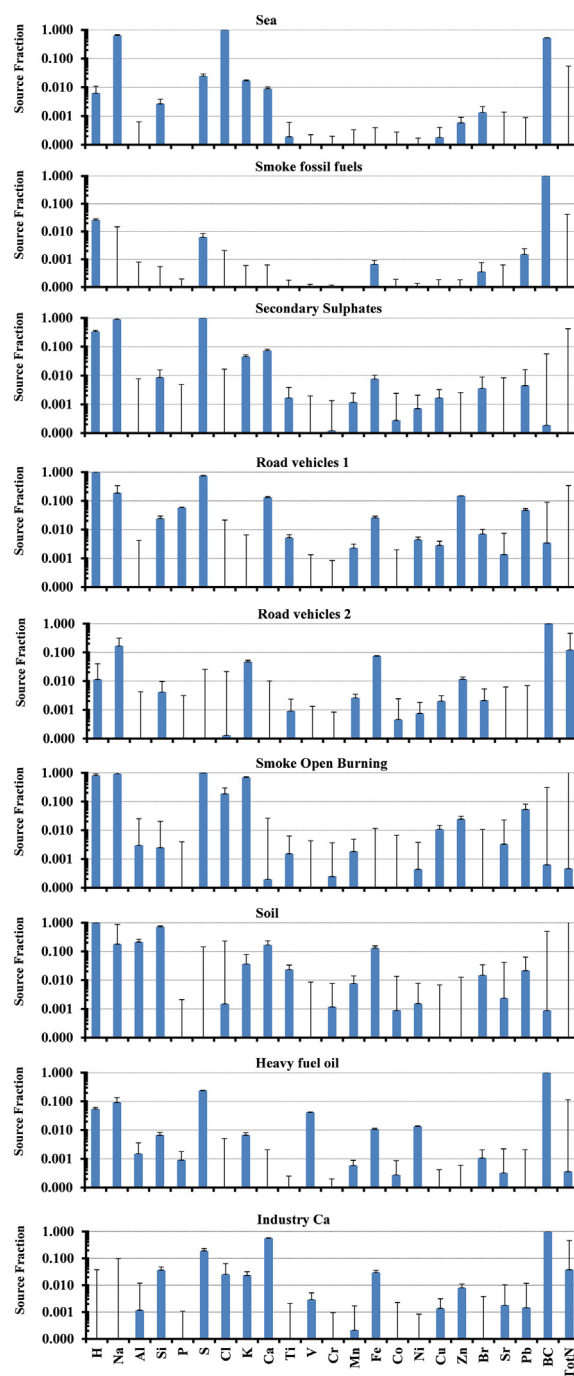


Fig. 2. Factors identified at Suva city, showing contribution of each factor to total PM_{2.5} mass. The vertical axis of each graph has been normalised so that the main element in each factor has a fractional contribution value of one.

contributions between the two models are included in health risk calculations (Supplementary Section B). The primary difference between the eight and nine factor PMF models is the inclusion of a second road vehicle factor. This extra factor, with $8 \pm 2\%$ contribution, is not formed by splitting the road vehicle factor, as it appears in the eight-factor model, but instead by re-distributing all elements, effectively reducing each of factors by a small percentage (Fig. 2, Supplementary Fig. A1). In terms of the contribution to total $PM_{2.5}$ mass, the ranking of sources from highest contribution to lowest contribution does not change between eight and nine factor models, except for slight variation in the smallest two factors. Percentage of total mass for each element, apportioned to each factor, is shown in Supplementary Fig. A2. These remain fairly consistent between the eight and nine factor models, although inclusion of the second road vehicle source factor in the nine factor model causes some redistribution of elements. Plots of the daily occurrence of each factor, by mass, show general similarity between eight and nine factor models (Supplementary Fig. A3 and percentage contribution Fig. A4).

More than 99% of the gravimetric mass was explained in the PMF model, with $R^2 = 0.99$ (Supplementary Fig. A5). This result represents a significant improvement on the particulate mass percentages able to be identified in previous studies (Garimella and Deo, 2007). In general, modelled PMF mass for each element show very good fit against IBA mass (Supplementary Figs. A6 and A7). The IBA v PMF fit for Cr and Co was poor compared to other species, due to the high instance of concentrations below the detection limit. Still, these results do not adversely affect the determination of source factors.

2.4. Meteorology

Meteorological data used in this study are based on hourly-averaged wind speed and direction data collected by the Australian Bureau of Meteorology (Australian Government, 2016) at the Suva city air sampling site. A summary of meteorological data, for the October 2014 to October 2015 period and their influence on $PM_{2.5}$ mass and BC concentration, are given in Isley et al. (2017). The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) model (Stein et al., 2015) has been used to calculate back trajectories. These have been calculated, commencing at three-hourly intervals, for a total of 24 h before arrival at the Suva City site, using an hourly timestep.

2.5. Risk analysis

Risk estimates were calculated for $PM_{2.5}$ and its components in Suva, using risk data from literature (included in Results). Mortality implications of risk were calculated according to a human health impact function (Cohen et al., 2004; Lelieveld et al., 2013; Fann, 2015; Broome et al., 2016; United States Environmental Protection Agency, 2017), using:

$$\Delta Y = Y_0 (1 - e^{-\beta \Delta PM}) \text{ Pop} \quad (1)$$

where:

- β mortality risk estimate as percentage increase
- Y_0 baseline mortality rate
- Pop exposed population
- ΔY mortality change (change in number of deaths expected per year)
- ΔPM change in concentration

To demonstrate this, mortality risk calculations for road vehicle PM have been included as Supplementary Section B. For Fiji, the baseline mortality rate is 8 per 1000 (Ministry of Health and Medical Services, 2015). The exposed population is 143,900 (Fiji Bureau of Statistics, 2007; World Population Review, 2017); being residents of Greater Suva (council areas of Suva, Lami, Nasinu and Nausori, Fig. 1) aged >30 years. This age group is selected as it relates most closely to

epidemiologically based risk data (World Health Organization, 2013a; Lelieveld et al., 2015; Broome et al., 2016).

3. Results

3.1. PMF data analysis

Elemental constituents of the source factors determined by PMF for the Suva $PM_{2.5}$ samples are shown in Table 1. Percentage mass contribution of each factor are separated for Sunday and mid-week samples as well as for wet and dry seasons. A comparison of the mass differences for each source factor, averaged over one year, is also shown by wet/dry season and week-day/Sunday.

In terms of the contribution to total $PM_{2.5}$ mass, the largest source is sea aerosol, followed by smoke from fossil fuels, secondary sulphates, road vehicles, smoke from open burning, windblown soil, industry Ca (an industrial source high in Ca) and heavy fuel oil. Back trajectory diagrams showing paths for air parcels 1 h prior to arrival in Suva city have been drawn for the peak 5% of occurrence of each source (Supplementary Fig. A8). These back-trajectories show that air has typically travelled mainly over open ocean in the hour prior to sampling in Suva. However for some sources (secondary sulphates, road vehicles, smoke from open burning, heavy fuel oil and industry Ca), these back-trajectories include airflow paths over Fijian land.

3.2. $PM_{2.5}$ source characteristics

The identifying features of each source factor are listed below. Wind conditions under which peaks of each factor occurred are shown in Fig. 3 and HYSPLIT back trajectories for each factor in Supplementary Fig. A8.

3.2.1. Sea aerosols

The largest factor identified in Suva's $PM_{2.5}$ was sea aerosol; comprising Cl, Na, BC, S, K, Ca, H, Si, Br and Ti (Fig. 2). The sea aerosol factor has a Cl/Na ratio of 1.56, representing fresh sea salt. Surface sea water has a Cl/Na, molar ratio of 1.16 (Keene et al., 1986), which is a mass ratio of 1.8. Whilst there is a slight loss of Cl compared with sea water; this factor has a similar Cl/Na ratio to sea aerosols measured at Australia's Cape Grim coastal air monitoring station (Cohen et al., 2014). Peak occurrence of the sea aerosol factor coincide with moderate winds (4 m/s to over 7 m/s), mainly from the southeast; being, on average, 11% higher than 2014–2015 wind speed averages for Suva. The presence of black carbon (BC) in this factor, accounting for approximately 16% of total BC (Supplementary Fig. A2), may show influence from shipping sources in the Pacific or land-based emissions in Suva, mixing with sea-spray. Still, this proportion of BC in sea aerosol is similar to Australian sites (Cohen et al., 2014). Long-range sources may also contribute to BC (Andreae, 1983; Liu et al., 2011).

3.2.2. Smoke from fossil fuels

The smoke from the fossil fuels factor is driven by BC and contains H and S, which are characteristic of smoke. As K is not present, this indicates an absence of biomass burning as a contributing source. Smoke from fossil fuels contributes 21% to total $PM_{2.5}$ mass and 57% of total BC mass (Supplementary Fig. A2). Given that coal burning does not occur in this region, this factor is likely to represent primarily diesel emissions.

Prevailing winds during peak detection days were from the south-east, as is typical for Suva, which would transport emissions from smaller shipping vessels off the coast of Fiji (discussed below). Yet, 49% of winds (peak detection days) are from the northeast quadrant. More than one-third of all wind speeds (peak detection days) were below 2 m/s; with the majority (70%) of low-speed winds occurring in the northeast quadrant. Relevantly, Suva's two thermal (diesel oil and

Table 1

Average contribution of each source factor identified to total PM_{2.5} mass; averaged over November 2014 to October 2015. Average factor contributions are also delineated by mid-week and Sunday results as well as by wet and dry season, including a comparison of average factor mass for each of these periods. Wet season results are for November through April and dry season results are for May through October. Midweek results primarily represent Wednesday samples, with the inclusion of eight samples from other week days. Industry Ca represents an industrial factor characterised by a high proportion of calcium.

	Percentage contribution to total PM _{2.5} mass					Mid-week factor mass/Sunday factor mass	Dry season factor mass/wet season factor mass
	All samples n = 92	Sundays n = 44	Mid-week n = 48	Wet season n = 52	Dry season n = 40		
Sea	25 ± 1	26 ± 3	22.40 ± 0.05	22 ± 3	26 ± 3	1.3	1.3
Smoke from fossil fuels	21 ± 2	21 ± 2	20.84 ± 0.04	25 ± 2	16 ± 2	1.5	0.7
Secondary sulphates	18 ± 1	22 ± 2	12.47 ± 0.03	18 ± 2	15 ± 2	0.8	0.9
Road vehicles 1	9 ± 1	9 ± 2	9.52 ± 0.03	10 ± 1	9 ± 2	1.7	1.0
Road vehicles 2	8 ± 2	4.7 ± 0.8	11.00 ± 0.02	6.2 ± 0.8	12 ± 2	3.5	2.0
Smoke from open burning	7 ± 1	4.9 ± 0.7	9.59 ± 0.02	8 ± 1	7 ± 1	2.9	1.0
Soil	5 ± 1	4.3 ± 0.6	6.86 ± 0.03	4.4 ± 0.7	7 ± 2	2.4	1.9
Heavy fuel oil	2.9 ± 0.7	3.6 ± 0.8	2.32 ± 0.01	3.3 ± 0.6	2 ± 1	1.0	0.8
Industry Ca	4 ± 1	4 ± 1	5.00 ± 0.02	3.6 ± 0.7	6 ± 1	1.8	1.8

residual oil) power generation plants are positioned to the northeast of the sampling area, at Kinoya (Fig. 1).

Contributions would also include industrial activity directly to the northeast of the sampling site (1 km), where the steel recycling mill, flour mill, brewery and several smaller industries operate. Under low-speed northeasterly winds, emissions from these industries and smoke from vehicles in Suva would not disperse readily and would therefore be likely to be detected more strongly at the sampling site. Peak concentrations of this factor occurred in April and May of 2015 (Fig. A3). When considered as a percentage of total PM_{2.5}, peak values for this factor are more evenly distributed across the year (Fig. A4).

As mentioned above, small shipping craft around Suva would contribute to this fossil fuel factor. Internationally, non-cargo vessels; passenger and fishing ships, tugboats and others; represent 50% of the shipping fleet (Eyring et al., 2005). Fuel use and accurate current

registration statistics for small shipping vessels in Pacific Island countries including Fiji are difficult to obtain (Holland et al., 2014), however observations of port activities during field sampling indicated that smaller vessels comprise around half of the shipping fleet in Suva. Holland et al. (2014) calculate, for Fiji, that shipping accounts for 22% of all fuel imports, representing up to 325 million litres of fuel annually; with the fleet being 'typically made up of old, poorly maintained fossil fuel [diesel] powered vessels' p. 94.

Fuel usage may also be higher than reported, as some vessels re-fuel outside of port and this is not included in national statistics (Eyring et al., 2005). These shipping emissions were not included in previous emission estimates for Suva (Isley et al., 2016), which provides some explanation for high ambient concentrations of black carbon by comparison with emissions estimates. Whilst larger ships tend to use heavy fuel oil, smaller ships use either marine diesel or automotive diesel.

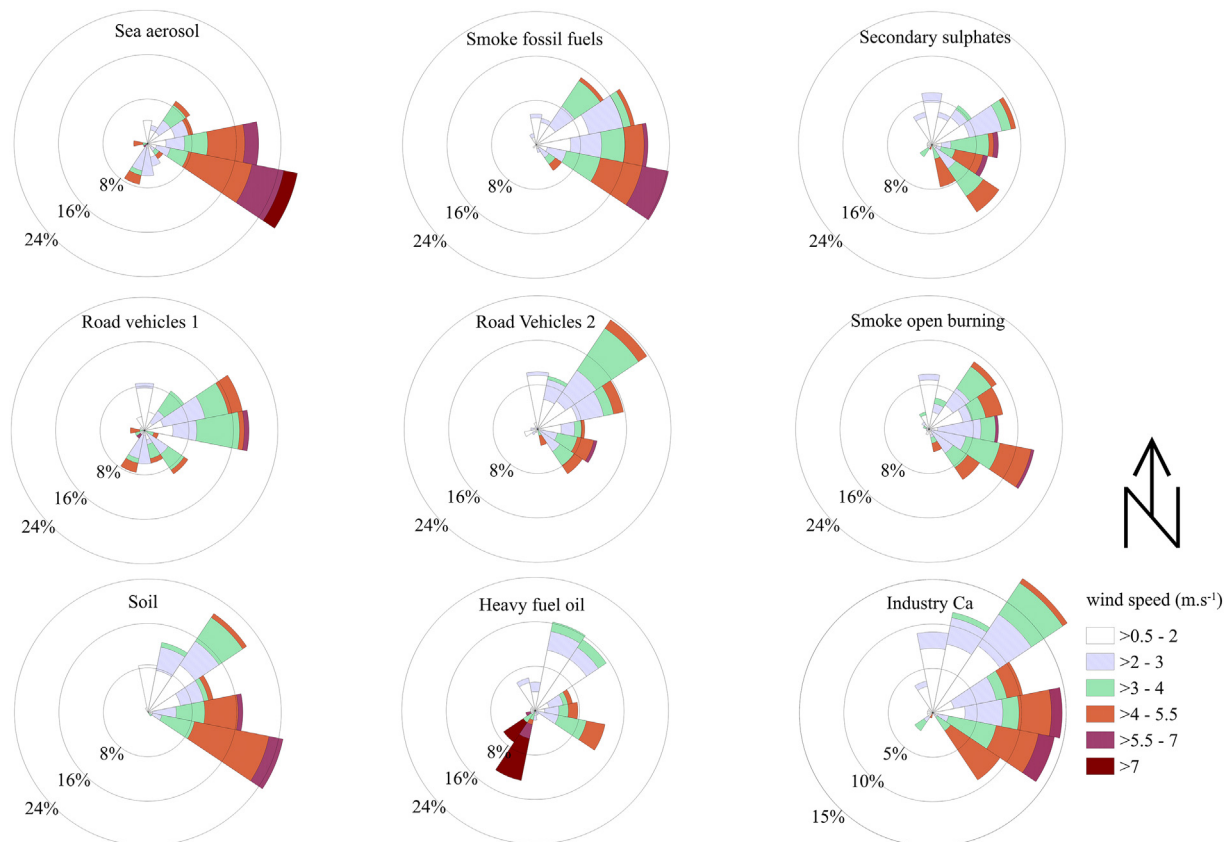


Fig. 3. Wind roses showing wind-speed and direction dependencies of source factors at Suva City. For each source, the percentage of counts for each wind direction and speed category are shown for peak 5% of occurrence of that source, based on the PMF model.

Marine diesel typically contains around 0.155% sulphur by mass (Kasper et al., 2007), over three times the sulphur content in Fiji's automotive diesel. Whilst marine diesel contains 50 times less sulphur than heavy fuel oil, particulate emissions were found to be only a factor of two lower (Winnes and Fridell, 2009). Marine diesel does not contain the nickel and vanadium characteristic of heavy fuel oil (Kasper et al., 2007), meaning that marine diesel would not appear in the 'heavy fuel oil' source, but would rather appear here in the 'smoke from fossil fuels' source.

Very small boats in Fiji, that have outboard motors, use the same petrol and diesel fuel as brushcutters, generators and motor vehicles (Holland et al., 2014). Petrol and diesel-fueled craft are also common in other Pacific Islands (Pacific Regional Data Repository, 2015). Because two-stroke outboard engines burn oil, they emit high levels of particles compared to other engine types, including car engines (Department of the Environment and Water Resources, 2007; Kasper et al., 2007). These very small boats are largely unregulated in Fiji.

3.2.3. Secondary sulphate

The secondary sulphate factor represents a composite of sulphates from natural marine sources and anthropogenic sources. Secondary sulphates form when SO_2 is scavenged by water droplets and is oxidised to sulphate, over a period of approximately one day, effectively mixing particles from multiple emission sources. Considering that air arrives in Suva after travelling hundreds of kilometers over the Pacific Ocean, spending only minutes, on average, over the land area of Suva (Isley et al., 2017), there is little time for emissions from land-based activities to be converted to sulphates. Ocean-derived biogenic sulphur, due to phytoplankton blooms, has been shown to contribute to inland aerosols (Gaston et al., 2010). This algal source is anticipated to contribute significantly to Suva's secondary sulphates. Whilst Suva does not experience highly distinct summer and winter seasons, the secondary sulphate factor displays seasonality (Supplementary Figs. A3 and A4). Peaks in the summer months (December to February) correspond to peak phytoplankton activity. Emissions from shipping in the Pacific, particularly the use of high-sulphur fuels, would also contribute to secondary sulphates. Back trajectory modelling (Supplementary Fig. A8), shows that some peak days for this factor correspond to periods when, quite unusually, the air mass has come from over the land area of Fiji. This indicates that local, human-influenced sources, such as industrial activity and vehicles, also contribute to secondary sulphates.

3.2.4. Road vehicles

The 'road vehicles 1' factor is driven by H and S, whereas for 'road vehicles 2', BC is the key component. Both of these factors contain elements relating to road-dust (Fe, Si and Ti), fuel and lubricant (Na, Zn, Br and Ca). The high sulphur in 'road vehicles 1' factor may indicate that it relates to vehicles using diesel fuels, containing up to 500 ppm sulphur. This would include most heavy vehicles such as trucks and buses. Moreover, the 'road vehicles 1' factor also shows higher proportions of road dust elements, consistent with the higher amounts of dust suspended with increasing vehicle mass (United States Environmental Protection Agency, 2006b). Peak levels of the 'road vehicles 1' factor occurred mainly under easterly and east-northeasterly conditions (Fig. 3); which is downwind of Suva's main bus stand.

The 'road vehicles 2' factor differs in that its peak occurrence relates to winds from the northeast, which contains the highly populated residential area of Suva; along with traffic corridors to the outer areas of Nasinu and Nausori (Fig. 1). Traffic emissions occur over a wide area of Suva and a lack of detailed traffic data in Fiji make it difficult to characterise vehicle emissions comprehensively. The presence of Pb in the 'road vehicle 1' factor may be due to brake wear (Pant and Harrison, 2013), or resuspension of roadside dust; which has been shown to contain up to 5161 mg/kg (Department of Environment Fiji, 2007).

3.2.5. Smoke from open burning

Smoke from the open burning factor is differentiated from the fossil fuel smoke factor by the high K content, indicative of biomass burning. Open burning of wastes is practiced widely across the Pacific Islands. In Suva, more than half of households burn a portion of household waste and greenwaste (Isley et al., 2016). Although waste burning activities are more concentrated in residential areas, the data shows that this source factor represents $7 \pm 1\%$ of $\text{PM}_{2.5}$ mass measured in Suva's city centre. It is not surprising therefore, that peak occurrence of this factor coincides with a high proportion of winds emanating from the residential areas in the northeast sector of Suva (Fig. 3). Back-trajectories for this factor (Supplementary Fig. A8) also include air parcels originating from northeasterly areas of Suva in the hour prior to sampling.

3.2.6. Soil

A Soil factor is characterised by Si, Al, Na, H, K, Ca and total N. This factor contains contributions from windblown soils, including dust emissions due to vehicles on unsealed roads and agricultural disturbance of soils. Peak detections of this factor were associated with southeasterly winds over 4 m/s. This is not surprising, as soil erosion potential of exposed areas increases rapidly with increasing wind speed (United States Environmental Protection Agency, 2006a). The soil factor is also related to northeasterly winds from the inland areas of Suva (Fig. 3).

3.2.7. Industry Ca

A source that was characterised by a high calcium content, labelled 'industry Ca' appears to represent a combination of emissions from cement industry operations at Lami and Nasinu and quarrying activities at Nasinu (Fig. 1). Several smaller cement industry operations also occur throughout the Suva area. Peak concentrations of this source factor occurred predominately under northeasterly wind conditions, from the direction of Nasinu (Fig. 3). Back trajectories (Supplementary Fig. A8) show also that air parcels originate from the northwest where larger-scale concrete industry operations are present in Lami.

3.2.8. Heavy fuel oil

A source factor representative of heavy fuel oil emissions was also identified, primarily due to the V/Ni ratio of 3.13, which is consistent with heavy fuel oil sources (Viana et al., 2014). This source factor is typically referred to as 'shipping' emissions by many authors (Pandolfi et al., 2011; Kuang et al., 2015), as it represents emissions from large ships that typically combust heavy fuel oil. The use of diesel fuels other than heavy fuel oil by many ships in Fiji means that a label of 'shipping' from this source would be an under-representation of the total impact of shipping on Suva's $\text{PM}_{2.5}$. Peak values for this factor were associated with an unusually large proportion of winds from the southwest, the location of Suva Harbour.

3.3. Implications for mortality

A summary of the available literature addressing the health impacts of $\text{PM}_{2.5}$ components is provided in Table 2. This has been limited to studies published since 2014, as published reviews (included in Table 2) cover the period prior to 2014. From the studies presented, it is clear that $\text{PM}_{2.5}$ composition plays an important role in the resultant health effects from exposure. Oil burning, vehicle-related PM, various metals and BC/EC are amongst the $\text{PM}_{2.5}$ components for which health impacts have most commonly been reported, however results have varied between studies. In summary of Table 2, the risk from $\text{PM}_{2.5}$ sources, relevant to Suva, is: sea aerosol – inconclusive; fossil fuels – ischemic heart disease, lung cancer, inflammatory response and DNA damage; secondary sulphates – respiratory disease, mortality; road vehicles – respiratory and cardiovascular risk, affects cellular function, mortality association; open burning – lung cancer risk; soil – inconclusive, though Si is associated with mortality; heavy fuel oil – cardio vascular hospitalisations and adverse impact on survival. Relationship of $\text{PM}_{2.5}$

Table 2Studies released in the last three years examining PM_{2.5} composition and health impacts.

Study	Type	Component of interest	Findings
Metals or specific elements/compounds Jia et al. (2017)	Review of cytotoxicity studies	Inorganic, organic, aqueous	Interaction between components affects toxicity. Associations are not clear and specific. Inorganic metals/non-metals: oxidative damage and cell viability reduction; transition metals via oxidative damage and inflammatory effects. Organic components is via inflammatory effects and genetic toxicity; aqueous compounds via cell viability reduction, DNA damage and apoptosis Lung tissue inflammatory response results were inconsistent.
Roper et al. (2017)	Toxicology	Metals	
Dai et al. (2014)	Epidemiology	Ni	Ambient Ni exposure is associated with increased blood pressure
Liu (2016)	Epidemiology	As and Cu Br, K, Na	Br, K, Na ⁺ , and sulphate associated with emergency department visits As and Cu associated with hospital admissions for stroke and pneumonia
Liu (2016)	Epidemiology	NH ₄ ⁺ , NO ₃ ⁻ , SO ₄ ²⁻ & V	Associated with increased mortality risk. Risk metrics not given.
Atkinson et al. (2015)	Meta-analysis	SO ₄ ²⁻ , NO ₃ ⁻ , EC, OC, particle number concentration (PNC) and metals	SO ₄ ²⁻ , NO ₃ ⁻ , EC and OC positively associated with all-cause, cardiovascular and respiratory mortality, most strongly for carbon; PNC: mainly positive associations, confidence intervals overlapped 0%; Metals: insufficient data; Suggests EC and secondary inorganic aerosols are associated with adverse health effects Associated with mortality (1.14; 95% CI: 1.06, 1.23 per 200 ng/m ³)
Beelen et al. (2015)	Cohort	S	
Chung et al. (2015)	Epidemiology	NH ₄ ⁺ , NO ₃ ⁻ , SO ₄ ²⁻ , EC, OC, Na, Si	Reduction in SO ₄ ²⁻ associated with increased life expectancy; reduction in NH ₄ ⁺ and Na ⁺ associated with increased life expectancy in nonurban counties only. SO ₄ ²⁻ and Na strengthen PM _{2.5} association with mortality.
Chung et al. (2015)	Epidemiology	EC, Si and NO ₃ ⁻	One standard deviation (SD) increase associated with 1.3% (EC, SD = 0.24 µg/m ³), 1.4% (Si, SD = 0.03 µg/m) and 1.2% (NO ₃ ⁻ , SD = 0.85 µg/m) increase in monthly mortality.
Gray et al. (2015)	Review epidemiology & toxicology studies	Metals	Suggest toxicity of Fe, Ni, V and Zn, however inconclusive. Health associations are more commonly reported for Fe, Ni, V and Zn than As, Cu, Mn and Se. Cu, Fe, Ni, V, Zn can produce inflammatory responses.
Peterson et al. (2015)	Cohort	Polycyclic aromatic hydrocarbons	Affects brain function in children
Wolf et al. (2015)	Cohort	Road dust, K, Si, Fe	Association with coronary events
Eeftens et al. (2014)	Cohort	Ni, S, Cu, Fe, K, Si, V, Zn	Ni and S associated with adverse lung effects, varied by cohort.
Wagner et al. (2014)	Toxicology	OC, non carbon components & trace elements	Negligible effects
Wang et al. (2014)	Epidemiology	Zn, Cu, Fe, Ni, Si, V	No association with cardiovascular mortality
Emission source-based studies			
Bind et al. (2016)	Epidemiology	Traffic-related PM _{2.5}	Increases risk of heart disease for those already in high-risk categories
Raspanti et al. (2016)	Epidemiology	Household biomass combustion	Chronic exposure is associated with increased lung cancer risk.
Samoli et al. (2016)	Epidemiology	Traffic emissions (PM ₁₀)	Association with cardiovascular admissions
Thurston et al. (2016)	Meta-analysis of cohort study	Windblown soil & biomass combustion	Not associated with IHD mortality
Hime et al. (2015)	Review of toxicology, epidemiology and clinical trial studies	Coal-fired power stations	Significant evidence of adverse health impacts both from primary and secondary particles. Some evidence that adverse impacts are worse than PM _{2.5} considered alone. Stronger associations with health outcomes for sulphate than PM _{2.5} .
		Road vehicles	Associated with mortality risk, cardiovascular and respiratory morbidity and adverse birth outcomes, exacerbates childhood asthma
		Domestic wood burning	Associated with adverse respiratory effects, particularly in children
		Forest fires	Increases respiratory morbidity, hospital admissions and asthma
		Crustal dust	Suggested associations with cardiovascular and respiratory effects. Severe dust storms are associated with increased daily mortality, but relationships are inconsistent.
		Biogenic	These have not been studied
		Sea salt	Some studies have demonstrated associations with mortality or respiratory effects, however results are inconsistent. Insufficient data.
		Organics	Insufficient direct evidence
		Sulphate	Associations have been observed, unclear if attributable to other components
		Nitrates	Evidence of respiratory effects, unclear if attributable to other components
Kiountzoglou et al. (2015)	Epidemiology	Ni, V, elemental carbon and sulphate	Higher (adverse) impact on survival in clusters with high Ni, V, EC & SO ₄ (fuel oil combustion & power plant emissions). Adults >65 years.
Kiountzoglou et al. (2015)	Epidemiology	Oceanic and crustal particles	Null or negative associations
Pun et al. (2015)	Epidemiology	Eight sources in PM ₁₀	Respiratory risk highest for vehicle exhaust, then secondary SO ₄ ; vehicle exhaust, regional combustion, secondary NO ₃ , aged (not fresh) sea salt and soil/road dust associated with respiratory hospitalisations.
Tong et al. (2015)	Toxicity	Traffic-related PM _{2.5}	Adversely affects cellular function
Bell et al. (2014)	Epidemiology	Road dust, Ca, BC, V, and Zn Road dust, sea salt, Al, Ca, Cl, BC, Ni, Si, Ti, and V	Associated with cardiovascular hospitalisations, people >65 years Associated with respiratory hospitalisations, people >65 years
Betha et al. (2014)	Health risk estimate	Biomass burning impacted aerosols	Higher lifetime carcinogenic risk than urban air
Dai et al. (2014)	Epidemiology	Sulphur	Short-term exposure associated with respiratory disease mortality — only one of 13

Table 2 (continued)

Study	Type	Component of interest	Findings
Grahame et al. (2014)	Review	BC mainly from diesel and traffic	species studied to give significant association BC from various sources is related to all cause, cardiovascular and lung cancer mortality
Grahame et al. (2014)	Review	Residential burning of biomass	Short-term exposure has adverse pulmonary effects and reduces immune defense; poor combustion (smouldering) relates to cytotoxic effects
Hennig et al. (2014)	Cohort	Traffic & industrial sources	Slightly stronger associations with cardiovascular disease but not significantly different from total PM _{2.5}
Li and Gibson (2014)	Risk modelling	Sulphur	Control of sulphur species from coal combustion has reduced deaths and hospitalisations
Lippmann (2014)	Review: toxicology & epidemiology	OC, EC, Cu from traffic and Ni and V from residual oil	These components are most closely associated with daily cardiovascular hospital admissions
Mauderly et al. (2014)	Clinical trial	Diesel, petrol, woodsmoke & coal emissions	Engine exhaust has greater respiratory and cardiovascular effects on animals than coal combustion or woodsmoke
Diesel emissions Mullins et al. (2016)	Toxicity	Canola biodiesel exhaust	Exposure elicits inflammation and reduces viability of human epithelial cell cultures
Thurston et al. (2016)	Meta-analysis of cohort study	Diesel exhaust/EC	Associated with ischemic heart disease mortality $3\% \pm 3\%/0.26 \mu\text{g}/\text{m}^3$ EC increase
Oeder et al. (2015)	Toxicology	Heavy fuel oil	Caused oxidative stress and inflammatory response in human lung cells
Oeder et al. (2015)	Toxicology	Diesel shipping fuel	Adversely influenced essential pathways of human lung cell metabolism, more strongly than heavy fuel oil
Ross et al. (2015)	Toxicity in calf thymus DNA	Diesel and biodiesel exhaust	Soy biodiesel emissions induced DNA damage
Bhavaraju et al. (2014)	Toxicity in rats lung tissue	Biodiesel low sulphur diesel	Both caused similar inflammatory response
Silverman et al. (2014)	Cohort	Diesel exhaust	Associated with lung cancer risk
Sun et al. (2014)	Review of cohort and case-control studies	Diesel exhaust	Exposure-response relationships remain unclear
Unosson et al. (2014)	Clinical trial	Diesel and biodiesel	Adverse vascular effects
Vermeulen et al. (2014)	Meta-regression of cohort studies	Diesel exhaust	Ambient and workplace concentrations pose substantial excess lifetime risks of lung cancer

sources to health impacts is a complex problem, due to the difference in PM_{2.5} mixtures from one location to another as well as findings that the toxicity of mixed PM_{2.5} components appears to differ greatly to that of individual components (Jia et al., 2017). Many studies in Table 2 relate to short-term exposures or specific demographic groups. For many sources or components, there is therefore insufficient information to make clear inferences about health risk (Wyzga and Rohr, 2015). Generalised risk estimates (long-term mortality) were reported for S, elemental carbon (EC), Si and NO₃⁻ and diesel EC (Table 2).

For determining the mortality implications of total PM_{2.5} concentration, a value of 1.06 (CI 1.04,1.08) per 10 $\mu\text{g}/\text{m}^3$ increase (Hoek et al., 2013) was used, as recommended by the World Health Organization (2013a) and used in (Broome et al., 2016). Hoek et al.'s (2013) risk estimate was based on a comprehensive review of risk assessment studies. Based on EC and BC studies, Hoek et al. (2013) estimated mortality risk of 1.06 (CI 1.05,1.07) per 1 $\mu\text{g}/\text{m}^3$ EC increase. Chung et al.'s (2015) epidemiological assessment using seven years of data from 518 PM_{2.5} monitoring stations in the United States of America (USA) estimated EC mortality risk at 1.013% (95% posterior interval (PI): 1.003, 1.022) per one standard deviation (SD = 0.24 $\mu\text{g}/\text{m}^3$) increase; consistent with Hoek et al. (2013). Measurements of BC and EC have been shown to be approximately comparable (BC:EC = 1 \pm 0.3) (United States Environmental Protection Agency, 2012). To account for this uncertainty in comparing EC and BC, mortality risk for Suva will be calculated using 1.045 (1.04 to 1.07) per 1 $\mu\text{g}/\text{m}^3$ BC increase. Further risk estimates are available for traffic related EC, S and Si. These risks were determined from only one study, in different settings to Suva, hence they must be used with caution. They will be considered only to explore potential risk in Suva. Thurston et al. (2016) followed 445,860 adults in 100 metropolitan areas for 23 years; diesel traffic related EC to be associated with mortality (risk = 1.03; CI: 1.00, 1.06 per 0.26 $\mu\text{g}/\text{m}^3$ EC increase); a rate of 1.021 (1.00, 1.06) per 0.26 $\mu\text{g}/\text{m}^3$ BC was used to

account for any BC/EC differences. Beelen et al.'s (2015) study of 19 European cohorts, determined risk from PM_{2.5} sulphur (1.14; 95% CI: 1.06, 1.23 per 0.2 $\mu\text{g}/\text{m}^3$), robust to adjustment for other pollutants and PM_{2.5}. Chung et al. (2015) reported risk for Si (1.014, 95% PI: 1.006, 1.024 for 1 SD = 0.2 $\mu\text{g}/\text{m}^3$ increase).

Reducing PM_{2.5} concentration in Suva has the potential to reduce annual mortality. Potential mortality reductions for greater Suva (council areas Suva, Lami, Nasinu and Nausori), for age group >30 years (143,900 people), are listed in Table 3 and Fig. 4. Firstly, mortality reduction has been presented according to the health risk of PM_{2.5} mass; for total PM_{2.5} and then for each component of PM_{2.5}, as identified in the PMF modelling. No data are presented for sea salt as these natural emissions cannot be easily reduced. Following this, potential mortality reduction has been presented for risk data specific to sources or components of PM_{2.5}; BC, traffic-related BC, S and Si. Major sources contributing to BC, S and Si have also been considered separately. Calculations have been based on the average of Wednesday and Sunday PM_{2.5} concentrations for 2014–2015, 7.4 $\mu\text{g}/\text{m}^3$. This may underestimate PM_{2.5} concentration and hence mortality risk in Suva, as Sunday levels were 30% less than weekday levels and as PM_{2.5} concentrations reported for residential areas were higher than for the city (Isley et al., 2017).

4. Discussion

4.1. PM_{2.5} sources

Modelling of source factors for the Suva city site, under either model, shows that the majority of PM_{2.5} mass originates from human activity, particularly combustion activities. Of this, a large proportion of PM_{2.5} is from fossil fuel smoke, but apportionment to specific individual industrial emission sources is not possible. Vehicle emissions were

Table 3

Potential mortality reduction calculated for greater Suva, for ages >30 years, due to reduction in different PM_{2.5} sources. The upper section shows reductions calculated using risk data for total PM_{2.5}. Lower sections use risk data for PM_{2.5} components. Whilst 100% reduction of PM_{2.5} from a given source is unlikely, this is listed in order to demonstrate the comparative mortality risk of different PM_{2.5} components. Risks marked * are based on only one study and should be considered only for the purpose of exploring risk potential in Suva. Ranges shown in brackets relate to the 95% confidence interval from risk estimate data as well as uncertainty in PMF modelling. Calculations are shown in Supplementary Section B.

	Potential mortality reduction based on health risk estimate data		
	10% source reduction	25% source reduction	100% source reduction
Risk calculation based on health risk of total PM _{2.5} , applied to components according to their average proportion in total PM _{2.5}			
Total PM _{2.5}	6 (4–6)	12 (10–15)	
PM _{2.5} smoke fossil fuels	1 (1–2)	2 (2–3)	11 (9–12)
PM _{2.5} secondary sulphates	1 (1–2)	2 (2–3)	9 (7–11)
PM _{2.5} road vehicles	1 (0–1)	2 (1–3)	7 (4–12)
PM _{2.5} smoke open burning	1 (0–1)	1 (1–2)	4 (3–5)
PM _{2.5} soil	1 (0–1)	1 (1–2)	3 (2–4)
PM _{2.5} heavy fuel oil	1 (0–1)	1 (0–1)	2 (1–3)
PM _{2.5} industry Ca	0 (0–1)	0 (0–1)	2 (1–2)
Risk calculation based on health risk of BC, applied to each component by average BC mass			
Total BC	11 (10–17)	28 (25–43)	
BC road vehicles	2 (2–4)	5 (4–9)	20 (15–36)
BC fossil fuel smoke	7 (5–10)	17 (12–25)	59 (48–97)
BC sea aerosol	2 (2–3)	5 (4–7)	19 (16–32)
Risk calculation based on diesel traffic-related BC*			
Road vehicles BC	3 (0–12)	9 (0–30)	35 (0–114)
Risk calculation based on sulphur, applied to each component by average sulphur mass*			
Total sulphur	16 (7–25)	39 (17–66)	
S from secondary sulphates	10 (4–16)	25 (10–43)	94 (39–164)
S from Smoke open burning	2 (1–5)	7 (2–11)	25 (11–45)
S from road vehicles	2 (1–5)	7 (2–11)	26 (8–52)
Risk calculation based on silicon*			
Total Si	2 (1–3)	3 (2–76)	
Si from soil	2 (1–3)	3 (2–5)	12 (5–22)

identified in distinct road vehicle factors, in addition to the fossil fuel factor. The combined contribution of road vehicle factors in the nine factor model, at 17% of PM_{2.5} mass, is lower than the 25% global average for traffic contributions (Karagulian et al., 2015). Given that vehicle

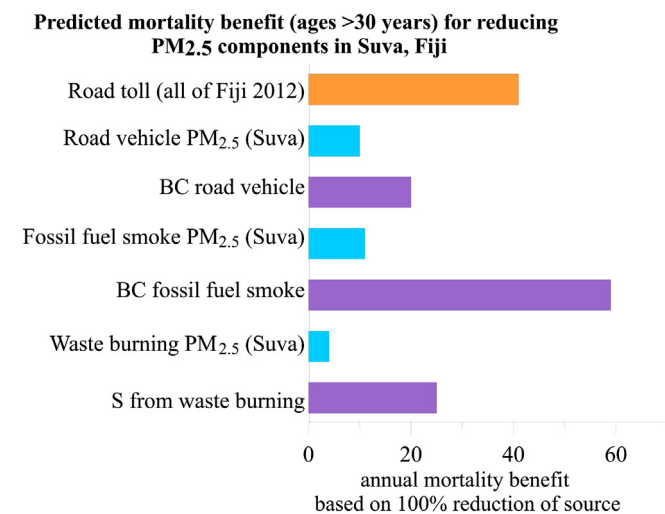


Fig. 4. Comparison of potential annual mortality reduction from reduction of different PM_{2.5} sources in Suva Fiji. These potential benefits are shown in terms of risk based on PM_{2.5} mass alone and risk based on specific PM_{2.5} components. These are compared to Fiji's road toll in order to demonstrate significance of each source.

registrations are increasing across Fiji and the Pacific Island Countries (Thoma, 2014; Fiji Bureau of Statistics, 2017); with Solomon Islands reporting over 60% increase in three years (World Health Organization, 2013b; World Health Organization, 2016) measures to control this source are becoming increasingly important.

Suva's PM_{2.5} sources varied from weekday to weekend. The mass attributed to the 'road vehicles 2' factor and to the open burning factor were around three times higher for midweek samples compared with weekend (Sunday) samples. A number of other sources; soil, industry Ca, road vehicles 1 and smoke from fossil fuels also showed higher mass midweek (1.5 to 2.4 times higher) than on Sundays. Very limited commercial and business activities are conducted on Sundays in Suva and so it follows that there are less vehicle movements to create exhaust emissions and suspended road dust; and less industrial or vehicle emissions contributing to the 'smoke from fossil fuels' factor. Further, agricultural and industrial operations, related to the soil and industry Ca sources, would also be reduced on Sundays. The smoke from open burning factor also shows reduced contribution on Sundays, which is consistent with this it being reserved as a church day and day of rest for many in Fiji, resulting in less clearing and burning of yard waste. By contrast, sea aerosols, secondary sulphates and heavy fuel oil factors were similar on weekdays and Sundays, with ratios ranging from 0.8 to 1.3. Ocean aerosols represent a natural consistent source not dependent on anthropogenic processes. Similarly, secondary sulphates and heavy fuel oil emissions are expected to remain similar across the week with ships working to move goods and passengers regardless of the day.

Source contributions also varied by season. Road vehicles 2, soil and industry Ca factors were approximately double, by mass, during the dry season compared to wet season. Dust emissions are related to moisture contents of exposed areas and therefore in the dry season dust emissions from unsealed roads and work yards, exposed areas, agricultural and industrial activities are increased (United States Environmental Protection Agency, 2006b). Likewise for Industry Ca, particle emissions rates for concrete manufacturing processes and quarrying activities are subject to the moisture content of material being handled (Department of the Environment and Energy, 2008; Department of Sustainability Environment Water Population and Communities, 2012).

4.2. Implications for mortality

Potential mortality impact for Suva, based on PM_{2.5} mass, represents a rate of 0.34/1000 persons (Table 3, mortality estimate divided by Suva's population of 330,000). Lelieveld et al. (2015) estimated mortality based on PM_{2.5} mass in the Western Pacific (including Pacific Islands and China) to be 0.85/1000; projected to increase to 1.2/1000 if air pollution sources were not addressed. By comparison, Australia had an estimated 0.2/1000 deaths attributable to urban air pollution (Australian Government Institute of Health and Welfare, 2016; Australian Bureau of Statistics, 2017). Estimates based on non-specific PM_{2.5} mass may underestimate mortality, especially where, as in Suva, carbon compounds contribute strongly to PM_{2.5} (Lelieveld et al., 2015). When toxicity of components is considered, potential mortality may be up to 0.67/1000 for Suva.

Mortality risk of BC from road vehicles could be responsible for around 20 deaths per year in Suva, or higher if using risk ratios for traffic-related BC. Smoke from fossil fuels has an even higher impact on mortality predictions; with predictions (Table 3) exceeding the nation's road toll of 41 in 2012 (Fiji Land Transport Authority, 2013). Fossil fuel smoke sources implicated from analysis of wind conditions (Section 3.1, Fig. 3) are shipping (diesel), power generation (diesel and residual oil) and diesel burning by Walu Bay industries: steel recycling mill, port-related industries, flour mill, brewery and other small industries. Traffic emissions may also contribute to this fossil fuel smoke factor. The sea aerosol factor in Suva comprises an average of 0.4 µg/m³ BC, meaning that it also carries mortality risk. This is likely

to be largely due to shipping and long range BC transport (Section 3.1), making BC from this source difficult to reduce.

The potential risk of S in PM_{2.5} indicates that secondary sulphates, smoke from open burning and road vehicles represents the most significant mortality risk of the emission sources modelled. Secondary sulphates in Suva represent a composite of biogenic marine sources with shipping emissions and land-based combustion emissions (Section 3.1); the relative contributions of these sources are unknown. It is also not known how the source of secondary sulphates affects their toxicity. Biogenic marine sulphates are an unmodifiable risk, largely because they are naturally occurring. Yet, secondary sulphates from vehicles, industry, waste burning and shipping may be reduced. Based on health risk of S, smoke from open burning and road vehicles may together contribute to Suva's annual mortality at a rate equivalent to the national road toll. The PMF determination of source contributions indicates PM in the Suva City area; waste-burning contributions to PM_{2.5} are likely to be greater in residential areas. Considering that Suva has waste collection services, emissions from open burning of waste seem largely unnecessary and removal of this emission source could potentially reduce annual mortality between 4 (based on PM_{2.5} health risk) and 25 (based on sulphur health risk).

5. Summary & conclusions

The IBA and PMF techniques have been used to determine the source contributions to Suva's PM_{2.5}. Modelling of source factors, for the Suva city site, shows that the majority of PM_{2.5} mass originates from human activity; particularly combustion activities. A large proportion of PM_{2.5} mass originates from fossil fuel (diesel) smoke. Unlike natural emission sources, such as sea aerosol, which cannot be reduced; by targeting fossil fuel emissions and open burning, it is possible to reduce Suva's atmospheric PM_{2.5} loadings. Considering the differential mortality risk of the PM_{2.5} components BC and S, the greatest health benefit in terms of reduced mortality would be gained by reducing fossil fuel smoke, vehicle related emissions and smoke from open burning of wastes. Analysis of epidemiological associations between Fijian health statistics and air quality data are recommended to further understand the health risk of air pollutants in the local context. Ongoing sampling is also recommended in order to statistically improve the PMF analysis and to determine the effectiveness of emission reduction measures.

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Appendix A. Supplementary data

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Paper Five

Radiocarbon determination of fossil and contemporary carbon contribution to aerosol in the Pacific Islands

Isley C. F., Nelson P. F., Taylor M. P., Williams, A. A., Jacobsen. G. E. *Science of the Total Environment* 643:183-192.

Supplementary information in Appendix D

Whilst this paper also deals with source apportionment of air particulates, it specifically focusses on the carbon content. Paper One and Paper Four both indicated that Suva has black carbon concentrations that are higher than might be expected for a small isolated city on a tropical island. Hence carbon particles in Suva's air warranted further investigation.

In addition, the carbon measurements to date have depended upon a single method, the Laser Integrating Plate Method (Taha *et al.* 2007). As with any method, this has limitations, which are discussed in Paper One. This provides a further reason for this study, as here a thermal method is used to extract and measure the carbon content, providing a different measure of carbon particulates.

Apart from measuring carbon mass, this paper examines elemental and organic carbon fractions. Using accelerator mass spectrometry, the proportion of these fractions that were fossil-sourced and contemporary-sourced were determined.

This study supports the previous papers in finding that carbon levels in Suva's air are indeed of significance from a health viewpoint. Further to this, as for Paper Four, it shows that the carbon sources in Suva city area are dominated by fossil fuels (vehicles, industry). By contrast, the residential area studied showed a much higher proportion of contemporary carbon, which demonstrates the impact of biogenic emissions as well as open burning

practices in residential areas, for Suva; but which are typical throughout the Pacific Island region. The very low carbon levels at the coastal site studied provide further evidence to Paper One and Paper Two that air quality and exposure are highly affected by local emissions practices.



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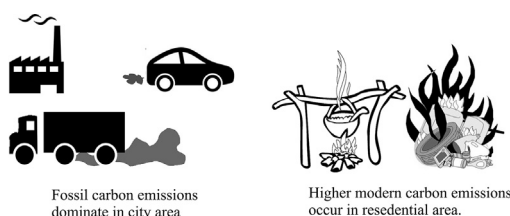
Radiocarbon determination of fossil and contemporary carbon contribution to aerosol in the Pacific Islands

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HIGHLIGHTS

- Aerosol elemental and organic carbon were determined for a Pacific Island location
- Fossil and modern components of carbon were quantified
- City areas were dominated by fossil carbon, from industry, vehicles and power generation
- Residential areas showed greater modern carbon, from biogenic emissions, waste burning and cooking
- Prior city-based studies have not captured the impact of open burning in residential areas

GRAPHICAL ABSTRACT



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Biogenic emissions

ABSTRACT

Combustion emissions are of growing concern across all Pacific Island Countries, which account for >10,000 km² of the earth's surface area; as for many other small island states globally. Apportioning emissions inputs for Suva, the largest Pacific Island city, will aid in development of emission reduction strategies. Total suspended particulate (TSP) and fine particulate (PM_{2.5}) samples were collected for Suva City, a residential area (Kinoya, TSP) and a mainly ocean-influenced site (Suva Point, TSP) from 2014 to 2015. Percentages of contemporary and fossil carbon were determined by radiocarbon analysis (accelerator mass spectrometry); for non carbonate carbon (NCC), elemental carbon (EC) and organic carbon (OC). Source contributions to particulate matter were identified and the accuracy of previous emissions inventory and source apportionment studies was evaluated. Suva Point NCC concentrations ($2.7 \pm 0.4 \mu\text{g}/\text{m}^3$) were four times lower than for City ($13 \pm 2 \mu\text{g}/\text{m}^3$ in TSP) and Kinoya ($13 \pm 1 \mu\text{g}/\text{m}^3$ in TSP); demonstrating the contribution of land-based emissions activities in city and residential areas. In Suva City, total NCC in air was 81% (79%–83%) fossil carbon, from vehicles, shipping, power generation and industry; whilst in the residential area, 48% (46%–50%) of total NCC was contemporary carbon; reflecting the higher incidence of biomass and waste burning and of cooking activities. Secondary organic fossil carbon sources contributed >36% of NCC mass at the city and >29% at Kinoya; with biogenic carbon being Kinoya's most significant source (approx. 30% of NCC mass). These results support the previous source apportionment studies for the city

Abbreviations: ANSTO, Australian Nuclear Science and Technology Organisation; ¹⁴C, carbon 14 isotope; δ_{blank} , correction factor for blanks; EC, elemental carbon; EC_{biomass}, elemental carbon from biomass burning; EC_{contemporary}, elemental carbon from contemporary sources; F_C, fraction contemporary carbon; F_{M(corrected)}, corrected fraction of modern carbon; F_{M(measured)}, measured fraction of modern carbon; F_{M(blank)}, fraction of modern carbon in the blank; NCC, non carbonate carbon; NCC_{contemporary}, non carbonate carbon from contemporary sources; OC, organic carbon; OC_{biogenic}, organic carbon from biogenic sources; OC_{p_biomass}, organic carbon from primary biomass burning emissions; OC_{contemporary}, organic carbon from contemporary sources; OC_{p_fossil}, fossil organic carbon from primary sources; OC_{S_fossil}, organic carbon from secondary organic fossil sources; PM_{2.5}, particulate matter < 2.5 μm effective aerodynamic diameter; PNC, particle number concentration; SE, standard error; TSP, total suspended particulate.

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area; yet show that, in line with emissions inventory studies, biomass combustion contributes more PM_{2.5} mass in residential areas. Hence air quality management strategies need to target open burning activities as well as fossil fuel combustion.

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1. Introduction

There is evidence that combustion-related atmospheric particles (which are largely carbon) are more hazardous than for particles from other sources (Hoek et al., 2013; Betha et al., 2014; Grahame et al., 2014; Lippmann, 2014; Mauderly et al., 2014; Vermeulen et al., 2014; Chung et al., 2015; Hime et al., 2015; Kioumourtoglou et al., 2015; Oeder et al., 2015; Raspanti et al., 2016; Samoli et al., 2016). Diesel exhaust has been classified by the International Agency for Research on Cancer (2012) as carcinogenic to humans. The World Health Organisation (Janssen et al., 2011; Janssen and WHO, 2012) suggests the use of carbon as an indicator in reducing population exposure to combustion particulate matter. Shindell et al. (2012) argue that in addition to health benefits, reducing black carbon in the atmosphere also mitigates climate change impacts.

Concern regarding combustion emissions is common across the Pacific Islands, particularly those due to diesel fuels (Gleye, 2010; Dornan and Jotzo, 2012; Pacific Energy Summit, 2013; Escoffier et al., 2016; Taibi et al., 2016) and from waste burning (Thaman et al., 2003; Periathamby et al., 2009; Mataka, 2011; Owens et al., 2011; Woodruff, 2014; Mohee et al., 2015). This study has relevance not only for Fiji and other Pacific Islands, but for other small island states affected by open burning of waste (Mohee et al., 2015; Marra, 2016; Riquelme et al., 2016) and reliance on diesel combustion (Mishra et al., 2009; Lucas et al., 2017). Indeed, these combustion sources are significant issues globally for cities in developing countries (Guerrero et al., 2013; Mitra, 2014; The World Bank, 2014; Wiedinmyer et al., 2014).

Studies in Suva, Fiji (Isley et al., 2017a) indicate significant black carbon concentrations ($2.2 \pm 0.1 \mu\text{g m}^{-3}$ annual mean). Source apportionment studies for Suva (Isley et al., 2018a; Isley et al., 2018b) have been based, in part, on black carbon concentration determined by the laser integrating plate method (Taha et al., 2007). Variability in the composition of atmospheric particles may affect black carbon measurements (Janssen et al., 2012). Here carbon content is tested using a thermal method, to verify previous results. Elemental (EC) and organic carbon (OC) components are also quantified, as these are useful for source apportionment (Zheng et al., 2006; Zhang et al., 2012; Bernardoni et al., 2013; Liu et al., 2014).

Further, radiocarbon analysis allows identification of fossil and contemporary carbon components (Zencak et al., 2007). Radiocarbon analysis is a method widely used in scientific studies and by environmental regulatory authorities to determine biomass burning contributions (Sheesley et al., 2009; Department for Environment Food and Rural Affairs, 2012; Belis et al., 2014). The 5730 yr. half-life of ¹⁴C makes it an ideal tracer for identifying combustion products derived from fossil fuels (¹⁴C-free) versus those from biomass (Reddy et al., 2002). In contrast to elemental and molecular tracers; e.g., potassium and chlorine, methyl chloride and levoglucosan, which may exhibit high emission variability, radiocarbon (¹⁴C) measurements provide unambiguous source apportionment (Currie, 2000) of contemporary and fossil carbon aerosol, since it retains its identity throughout atmospheric chemical changes (Krecl et al., 2008). Use of ethanol from biomass sources in motor vehicle fuels can complicate this, however ethanol was not being added to Fijian motor vehicle fuels at the time of the air sampling (World Data Atlas, 2018).

Fijian source-apportionment studies (Isley et al., 2018b) have focussed on the inner city and indicate that diesel fuels are the most significant anthropogenic source. Emissions inventory studies (Isley et al., 2016) estimate wood and waste burning to contribute 35% of

PM_{2.5} (particulate matter < 2.5 μm effective aerodynamic diameter) emissions in the greater Suva area. Conversely, Suva City air sampling results (Isley et al., 2018b) show open burning to only represent 8% of PM_{2.5} mass. In south Asia, Gustafsson et al. (2009) noted a similar difference between the particle emissions inventory and elemental composition studies, which used black carbon. Interestingly, radiocarbon analyses showed a larger contribution from biomass combustion (Gustafsson et al., 2009). Accurate information on biomass contributions is necessary for effective air quality management. It was surmised (Isley et al., 2018b) that open burning source contributions to atmospheric particulate may be higher in Suva's residential areas, where open burning of waste is common practice (McDowall, 2005).

In this study, using the radiocarbon method, fossil and contemporary carbon ratios were determined for Suva, as well as whether these are consistent between inner city, residential and background locations. These data can be used to identify source contributions to total particulate concentration; evaluate the accuracy of emissions inventory and previous source apportionment studies and provide a solid scientific basis for the development of effective pollutant control measures.

2. Method

2.1. Locations

Airborne particulate samples were collected at three ambient monitoring sites in Suva: Suva City, Kinoya and Suva Point as shown in Fig. 1. The Suva bus terminal, city markets, an industrial precinct and shipping port activities all lay within 1 km of the Suva City site. Kinoya lies within Nasinu, Fiji's most densely populated area, containing a mixture of formal and informal (squatter) settlements. Suva Point is a mainly ocean-influenced site, with winds mainly from the southeast (Isley et al., 2017a); hence less influenced by city emissions than the other locations.

2.2. Sampling

Samples were collected during intensive sampling campaigns in October 2014, April to May 2015 and October 2015, as detailed in Supplementary Table S1. PM_{2.5} samples were collected at Suva City using an Aerosol Sampling Program (ASP) cyclone sampler (Cohen, 1996) operating at 22 L/min flowrate. Total particulate samples, at Suva City, Kinoya and Suva Point, were collected using a low-volume Ecotech Microvol sampler with flowrate 3 L/s (Australian/New Zealand Standard, 2006). In each case, quartz fibre filters were used (47 mm for TSP, 15 mm for PM_{2.5}), precleaned at 900 °C to ensure carbon contaminants were removed. Samples were chilled during transport to prevent volatilisation of organic carbon components.

2.3. Sample preparation

Sixteen air particulate filter samples were prepared. Each sample represents an average of 3 days of sampling for PM_{2.5} samples (City, 5 samples) and 16 days for TSP samples (all sites, 11 samples). Samples were placed in a desiccator with 30 mL of concentrated HCl for 24 h and exposed to HCl fumes to remove carbonates (Jordan et al., 2006). All remaining carbon, which includes organic carbon (OC) and elemental carbon (EC), is termed total "non carbonate" carbon (NCC) in this paper. Twelve of the samples were divided into half, one half to be analysed for NCC and the other for EC. These fractions were prepared as shown in Supplementary Fig. S1.

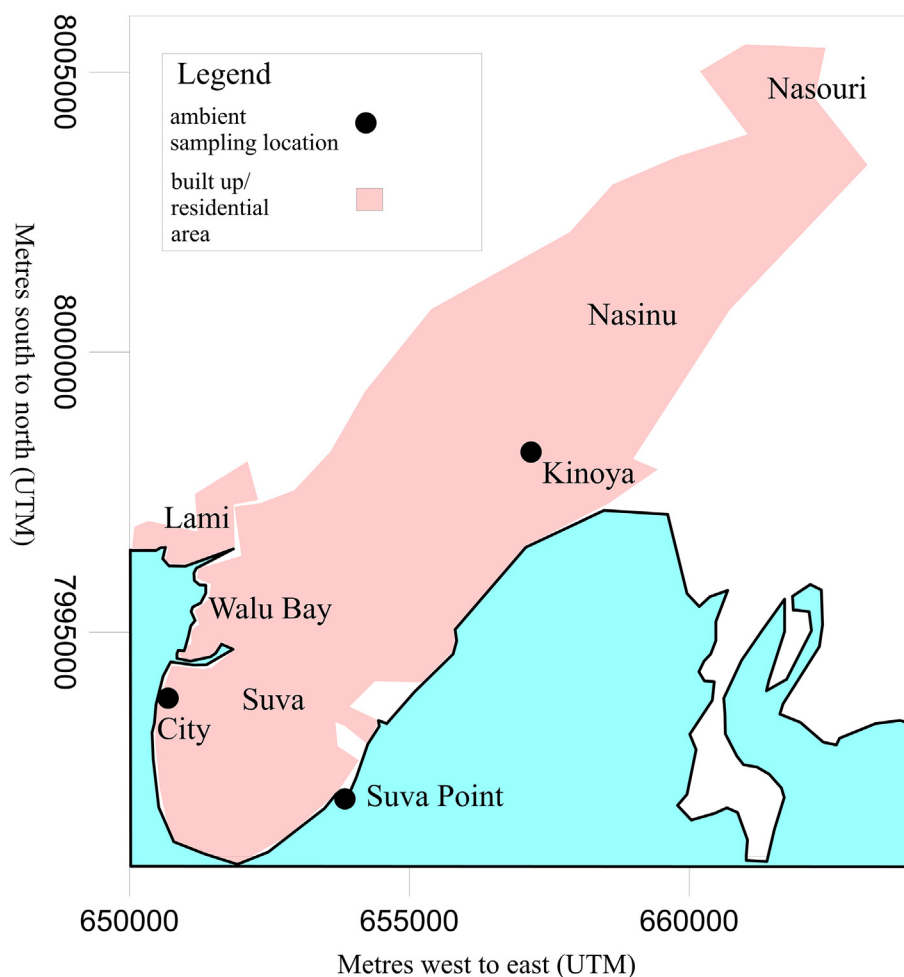


Fig. 1. Sampling locations for carbon aerosol sampling in Suva, Fiji, coordinates in Universal Transverse Mercator (UTM).

2.3.1. Total NCC

The first half of samples were cut into thin strips and packed into pre-cleaned quartz combustion tubes. The NCC procedure, presented briefly here, is detailed in Trumbore et al. (2016). Samples were heated to 900 °C in the presence of pre-cleaned CuO and Ag and held for 2 h at that temperature. Samples were then cooled to 600 °C and held for 2 h to allow any excess oxygen to recombine with the Cu₂O present and any nitrogen oxides that have formed to break down on the Ag wire catalyst, to nitrogen gas. Samples were then cooled to room temperature. Evolved CO₂ was cryogenically isolated using ethanol-dry ice and liquid N₂ traps, and quantified by manometry. CO₂ was reduced to graphite at 600 °C using hydrogen and an iron catalyst (hydrogen used at 2.5 times the carbon dioxide present).

Percentages of modern and fossil carbon (Stuiver and Polach, 1977) were determined by radiocarbon analysis (accelerator mass spectrometry) at the Australian Nuclear Science and Technology Organisation (ANSTO, Fink et al. (2004)). Sample volumes were used to calculate carbon concentration in air.

2.3.2. EC fraction

This method (Szidat et al., 2013) oxidises the OC fraction and removes it, leaving the refractory EC component behind for analysis. The remaining half of each sample was also cut into strips and loaded into a combustion tube. Air was evacuated from each tube, back filled with high purity oxygen (~700 mbar) and combusted at 375 °C for 1 h. Tubes were then cooled and they were evacuated a second time, filled with high purity He, heated at 580 °C (in line with the

IMPROVE-A protocol (Chow et al., 2007)) for 10 min and cooled to ambient temperature. Tubes were evacuated a third time, flame sealed and then subjected to the same treatment and analyses as NCC samples. The OC fraction was calculated by subtracting the EC mass from NCC mass.

Due to lower than anticipated carbon concentrations (based on black carbon concentrations, see Supplementary Table S2 and Isley et al. (2017b)), Suva Point samples and one Kinoya sample were only analysed for NCC. This was to ensure sufficient carbon sample mass for radiocarbon analyses. Whilst only two samples were analysed for Suva Point, these represent a sampling period of 58 days over the October 2014–October 2015 period, with filters being combined in order to provide sufficient carbon for analysis. Radiocarbon results are included as Supplementary Table S3 and S4.

During thermal analysis, a fraction of OC undergoes charring to form pyrolytically generated EC, meaning that true EC concentrations can be overestimated or underestimated depending on the method and temperature protocol (Yang and Yu, 2002; Zhang et al., 2012). The oxygen combustion step (Cachier et al., 1989a) used here, reduces formation of pyrolytically generated EC to 10% or less of the original organic content (Cachier et al., 1989b); this potential error has been used in presentation of results. Conversely, the oxygen combustion step may remove substantial amounts of non-refractory EC, leading to underestimation of true EC mass (Zhang et al., 2012). EC from wood-burning has been shown to be the least refractory EC fraction (Elmqvist et al., 2006) and biomass burning is an important source in Suva (Section 1). Hence, this could result in underestimation of EC; although using a temperature of 375 °C here, as opposed to a higher temperature, should minimize this EC loss (Zhang et al., 2012).

2.4. Quality control

Two sets of duplicate samples were collected to allow ^{14}C concentrations to be defined with greater certainty. These were collected by placing Microvol samplers side-by-side at Kinoya and Suva Point. One Suva Point duplicate was destroyed during analysis (Supplementary Table S1). Additional part-samples were also destroyed, as noted in Table S1. Duplicate samples from Kinoya (Kinoya_TSP_3 and Kinoya_TSP_4) show a 3% difference in ambient NCC concentration, which is a 1.2% difference in modern carbon percentage. These differences are smaller than the variations between samples collected on different dates ($\text{SE} = 8\%$ for ambient NCC concentration and 2% difference in contemporary carbon percentage of NCC, Supplementary Table S3). For EC, the variation in duplicates was larger (8% of ambient EC mass concentration); this is likely due to the much smaller sample mass available for EC. Variation in carbon mass of City samples is similar to the variation seen in black carbon concentration results (in $\text{PM}_{2.5}$) for the same site (Isley et al., 2017a).

Carbon contamination introduced during storage, transport by air and road and sample preparation was determined by analysing two pre-cleaned unused filters (field blanks); both for NCC and EC. Five blanks and five standards were used to assess ^{14}C contamination introduced throughout the process. A blank, fossil carbon source (graphite) and a modern carbon standard (Oxalic acid, NIST SRM 4990C), at sizes comparable to the Suva samples, were added to pre-cleaned filters, processed and analysed; as described in detail by Dusek et al. (2014). The radiocarbon content of the samples ($f_{\text{M(measured)}}$) was corrected for blanks, and a subsequent correction was made for both the mass and f_{M} of filter blanks according to Eqs. (1) and (2) (Klinedinst and Currie, 1999; Jordan et al., 2006).

$$F_{\text{M(corrected)}} = [F_{\text{M(measured)}} - (\emptyset_{\text{blank}} \times F_{\text{M(blank)}})] / [1 - \emptyset_{\text{blank}}] \quad (1)$$

$$\emptyset_{\text{blank}} = \text{mass blank carbon} / \text{total mass carbon} \quad (2)$$

On average, blanks contained 30 μg NCC, which comprised 12.5% modern carbon. For EC samples, blank filters contained insufficient carbon (3 μg EC) for determination of the modern fraction. Error in determining modern carbon in EC was therefore assumed to be similar (on a percentage mass basis) to that in the NCC samples.

2.4.1. Using leaf samples to determine contemporary carbon

In order to ascertain the current ^{14}C content for vegetation around Fiji, five samples of leaves were also collected in April 2015 and analysed by AMS. In aerosol studies, the contemporary carbon fraction is defined as the ratio between the fraction of modern carbon in the sample and fraction of modern carbon found in short-lived materials (such as leaves) in the year of sampling (Stenström et al., 2011).

For Fiji, this is particularly important considering the ^{14}C impacts of nuclear testing in the Pacific (Andrews et al., 2016). Plant species for leaf, collected from the city area, Kinoya, Suva Point and a nearby forest area (Colo-i-Suva) were identified using Thaman (2012); the Fijian and botanical names of each plant are listed in Supplementary Section B. In order to ensure that leaf samples did not contain surface contaminants, each leaf was cleaned as described in Supplementary Section B.

The fraction of contemporary carbon in air particulate samples was equal to the fraction of modern carbon (F_{m}) of the PM sample divided by this F_{m} for leaf samples (Bench, 2004). For the five leaf samples, median F_{m} was 1.027, which is in agreement for the measured Southern Hemisphere atmospheric value of 1.0273 ± 0.0004 for 2015 (Nichol et al., 2018; Woods Hole Oceanographic Institution, 2018).

Atmospheric aerosols may originate from biomass which has accumulated ^{14}C over time, during which atmospheric ^{14}C values will have varied, particularly after commencement and then cessation of atmospheric nuclear testing from 1955 to 1963, which injected large amounts of ^{14}C into the atmosphere, nearly doubling the concentration

in the northern hemisphere and increasing this by approximately 60% in the southern hemisphere (Stenström et al., 2011; Hua et al., 2013). Using northern hemisphere atmospheric ^{14}C concentrations, Lewis et al. (2004) modelled a range of possible wood ^{14}C ratios for wood between 10 and 50 years old. Results gave a range between 1.1 and 1.3 and showed that 50-year old wood (harvested in 1999), has an F_{m} ratio of 1.3 Modern C. Using Lewis et al. (2004), Bench et al. (2007) determined that for combustion of wood approximately 50 years old (harvested in 1999), this could mean a 20% overestimation of contemporary carbon (1.3/the F_{m} ratio of the contemporary component). This assumption of wood combusted being 50 years old comes from Szidat et al. (2006)'s study of wood burning in Zurich, where wood is burned mainly for residential heating or for forestry waste burning. Much biomass burning in Suva is likely to be smaller branches or leaves and grasses, which would have ^{14}C concentrations closer to contemporary atmospheric values; yet burning of paper materials, particularly those using recycled paper, may emit carbon from older wood sources. As Fiji is in the southern hemisphere, this value for potential overestimation of contemporary carbon will be lower than determined by Lewis et al. (2004) and Bench et al. (2007), as the ^{14}C bomb peak did not reach the level it did in the northern hemisphere (Hua et al., 2013). A southern hemisphere study (Jordan et al., 2006) tested woodsmoke of trees and large branches of mixed sources and ages, providing a good indication of average ^{14}C concentrations in this region. Jordan et al. (2006) determined the F_{m} (corrected) of the woodsmoke to be 1.040 for NCC and 1.049 for EC and is the only study of this kind that the authors are aware of for the southern hemisphere. Using Jordan et al. (2006), the potential overestimation of contemporary carbon for Suva then becomes 1% ($F_{\text{m,woodsmoke}} / F_{\text{m,Suva,leaves}} = 1.040 / 1.027$) for NCC and 2% for EC.

2.4.2. Summary

Modern carbon percentage in NCC, determined by AMS, was corrected for blanks using Eq. (1). The contemporary fraction was then determined by dividing this by the F_{m} for leaf samples (1.027). Further to this, error bars are used to indicate the variation that may be due to the age of plant material at burning (Section 4.1, tabulated in Supplementary Tables S3 – S5). These same error metrics were applied for EC samples; as well as the error in determining the EC/OC fraction (Section 2.3.2).

To determine the contemporary fraction in organic carbon, the following mass equation was used; with the equivalent calculation for the fossil fraction (more detail in Supplementary Table S5):

$$\text{OC}_{\text{contemporary}} = \text{NCC}_{\text{contemporary}} - \text{EC}_{\text{contemporary}} \quad (3)$$

All error calculations from determination of EC and contemporary carbon fractions were propagated through OC calculations.

2.4.3. Additional data for comparison

To allow comparison with concurrent studies, samplers were located as close as possible to measurement of TSP, $\text{PM}_{2.5}$, particle number concentration and black carbon at all sites (Isley et al., 2017a; Isley et al., 2017b), summarised in Supplementary Table S2. $\text{PM}_{2.5}$ sources, determined from elemental concentrations, are available for Suva City (Isley et al., 2018b).

2.5. Estimation of sources

2.5.1. EC sources

Fossil EC sources provide a measure of fossil fuel combustion and contemporary EC of biomass burning, whether natural or deliberate. That is:

(Heal, 2014)

$$\text{EC} = \text{EC}_{\text{biomass}} + \text{EC}_{\text{fossil}} \quad (4)$$

2.5.2. OC sources

For OC, however, there are primary biomass, fossil and biogenic contributions and secondary biogenic and fossil contributions. (Heal, 2014)

$$OC = OC_{P_fossil} + OC_{P_biomass} + OC_{biogenic} + OC_{S_fossil} \quad (5)$$

OC_{P_fossil} is derived from EC_{fossil} using ratios from literature, summarised by (Gelencsér et al., 2007); with $OC_{P_fossil}/EC_{fossil} = 0.58$ (range 0.48–1.2)

$OC_{P_biomass}$ is also derived from ratios $OC_{P_biomass}/EC_{biomass} = 6.0$ (range 1.0–15.0) (Gelencsér et al., 2007).

The biogenic OC component ($OC_{biogenic}$) can then be determined by:

$$OC_{biogenic} = OC_{contemporary} - OC_{biomass} \quad (6)$$

The primary component of $OC_{biogenic}$ is determined from cellulose concentrations. As these were not measured, the $OC_{biogenic}$ component calculated here includes both primary and secondary biogenic OC.

Note that this equation from Heal (2014) does not allow for separate calculation of secondary organic carbon from biomass burning sources. Instead, this would be included within the calculated $OC_{biogenic}$.

The final component OC_{S_fossil} is derived from subtraction of the other OC components from total OC (Heal, 2014).

Considering that there are ranges of values or error margins applicable to each component in Eq. (5), a sensitivity analysis has been conducted. This entailed calculating the results for each component in Eq. (5) for scenarios with low and high values of EC, contemporary carbon, $OC_{P_fossil}/EC_{fossil}$ and $OC_{P_biomass}/EC_{biomass}$. These results are included as Supplementary Table S7.

3. Results and discussion

3.1. Total non carbonate carbon

Each site sampled shows distinct characteristics in terms of carbon mass and proportions of fossil and contemporary carbon. A summary of results, corrected for blanks is shown in Fig. 2. Results and calculations for individual samples are listed in Supplementary Table S3. City TSP results report NCC concentrations approximately double of those in $PM_{2.5}$ from the same site; showing that carbon particulate is not constrained to the $PM_{2.5}$ fraction (similarly, (Samara et al., 2014)).

Suva Point reported total NCC concentrations of $2.7 \pm 0.4 \mu g m^{-3}$; four to five times lower than Suva City and Kinoya (respectively). Prevailing winds in Suva during the sampling period were from the south-east (Isley et al., 2017a). Suva Point therefore receives little influence from land-based emissions, receiving winds mainly from the ocean. Concentration and composition of carbon at this site is therefore indicative of long-range transport.

Concurrent studies (Isley et al., 2018a) support this difference between the sites, reporting mean black carbon at Suva Point ($0.2 \pm 0.2 \mu g m^{-3}$) to be at least five times lower than for Suva City and Kinoya. Ultrafine particle concentrations (Isley et al., 2017b), another indicator of combustion emissions, similarly show a difference between Suva Point (mean particle number concentration (PNC) $1.48 \pm 0.04 \times 10^5 cm^{-3}$) compared to Kinoya (mean PNC $4.42 \pm 0.02 \times 10^5 cm^{-3}$) and Suva City (mean PNC $1.64 \pm 0.02 \times 10^6 cm^{-3}$); although ultrafine particle data represent only a brief sampling campaign. The higher carbon concentrations in Suva City and Kinoya indicate that local sources, such as vehicles, industry and open burning, contribute to Suva's carbon particulate.

Comparing NCC mass to total particulate and $PM_{2.5}$ mass (Supplementary Table S2), carbon particulate makes up a large proportion of

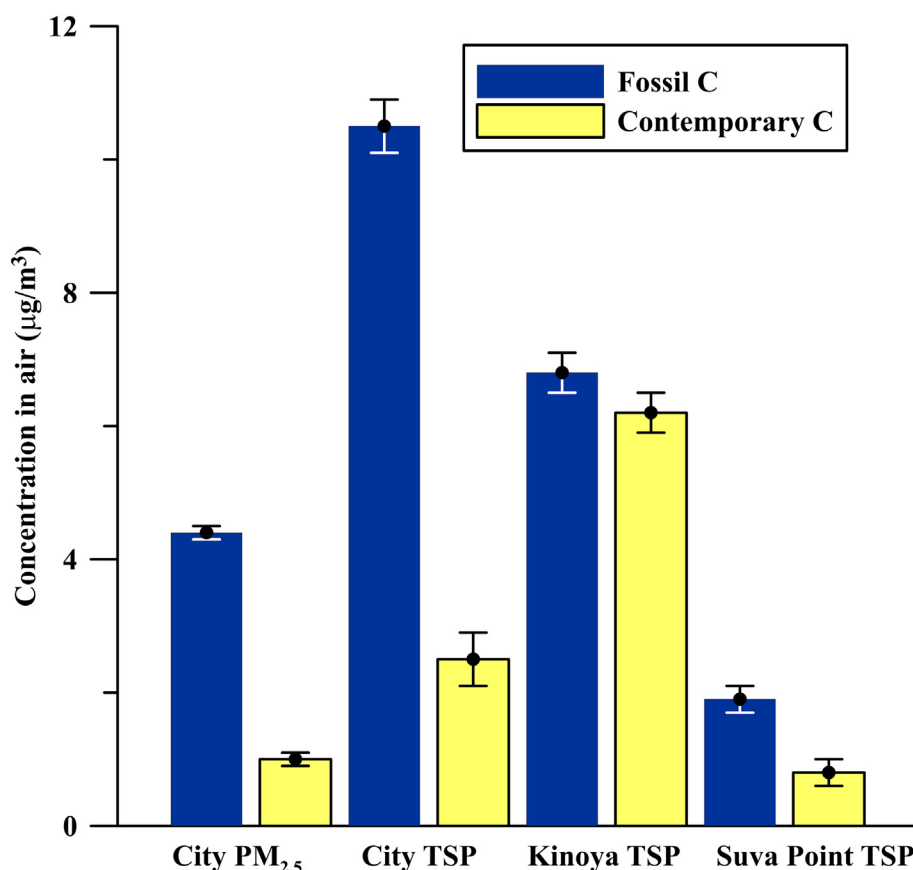


Fig. 2. Fossil and contemporary components of non carbonate carbon in $PM_{2.5}$ and TSP from Suva, Fiji. 2014–2015. Data for individual samples is included as Supplementary Table S3.

particulate mass in Suva. At Suva City, NCC accounts for around half of $PM_{2.5}$ mass and 45% of TSP mass. At Kinoya and Suva Point, NCC is equivalent to approximately 30% of TSP mass.

3.1.1. Fossil and contemporary components

Total NCC in air particulate samples from Suva City is dominated by fossil carbon (Fig. 4). The mean percentage of contemporary carbon of all City TSP and $PM_{2.5}$ samples is 19% (17%–21% of NCC). City TSP and $PM_{2.5}$ showed no significant difference between the percentage of contemporary carbon in NCC (Supplementary Table S3). The remainder (approx. 81%) of total NCC in Suva City's air is from fossil carbon sources. This is interesting, as globally, even in urban locations, contemporary carbon typically exceeds 30% of NCC mass (Heal, 2014). Fossil NCC accounts for approximately 45% of City $PM_{2.5}$ mass (Table S2, Table S3).

Kinoya samples (TSP) report, quite differently to the city, that 48% (46%–50%) of total NCC is from contemporary sources. This is typical of urban background locations globally (Heal, 2014). Kinoya contemporary NCC concentrations (6.2 (5.9 – 6.5) $\mu\text{g m}^{-3}$ in TSP) were more than double those in Suva City (2.5 (2.1 – 2.9) $\mu\text{g m}^{-3}$ in TSP). In Suva's residential areas, fires burning greenwaste and household wastes occur on a daily basis, with more than half of Suva households burning wastes (Isley et al., 2016). Household survey results for Suva (Isley et al., 2016) report that metals, glass and plastics are frequently removed from waste before burning, however only 10% of respondents removed paper, 16% removed plant materials and 28% removed food scraps before burning. Hence waste being burned is mainly composed of biomass materials. Biomass fuels are also used for cooking, with 20% of Suva residents using wood and 5% using coconut fibre (Isley et al., 2016).

Suva Point NCC samples contained 29% (23%–35%) contemporary carbon. Carbon detected at Suva Point may show some influence from local land-based activities, however, as discussed (Section 3.1), this site is more indicative of long-range sources. Interestingly whilst contemporary carbon sources, such as algal activity, would contribute

here, the bulk of carbon mass at this site (>70%) is of fossil origin; indicating contribution from ocean sources such as shipping, or other long-range transport of fossil carbon particulate.

3.2. Elemental and organic carbon

Organic carbon (Fig. 3) makes up the largest proportion of NCC at Suva City ($OC/EC = 2.5$ in $PM_{2.5}$, $OC/EC = 4$ in TSP) and Kinoya ($OC/EC = 5$). These results also match global data (Heal, 2014) in that contemporary carbon mass in EC is less than contemporary carbon mass in OC.

Suva Point samples yielded insufficient carbon for EC and OC determination, yet some information can be inferred through comparison with other studies on this site. Black carbon measured at Suva Point ($0.2 \pm 0.2 \mu\text{g m}^{-3}$ in $PM_{2.5}$, (Isley et al., 2018a) mean 9 sampling days over 9 months) was small compared to the NCC at Suva Point ($2.7 \pm 0.4 \mu\text{g m}^{-3}$ in TSP mean 58 sampling days over the same 9 months). However, different particle fractions (TSP v. $PM_{2.5}$) were considered in these two studies. Also, black carbon concentrations typically approximate EC concentrations (United States Environmental Protection Agency, 2012), not representing the OC component. Analysis of additional samples for this Suva Point site, allowing for determination of EC and OC, would provide more accurate information regarding composition and carbon sources.

Particulate ($PM_{2.5}$) composition at Suva Point was estimated (Isley et al., 2018a) from concentrations of 22 elements, using the method of Malm et al. (1994) and Cohen (2010). Only trace contributions (1%) of biomass smoke and black carbon (4%) were estimated; with the remainder being mainly salt (31%), sulphate (18%), soil and trace metals (14%), with 32% of the mass ($5.5 \pm 0.8 \mu\text{g/m}^3$) remaining undetermined (organics, nitrates and water vapour were not analysed). It is not known what proportion of the $2.7 \pm 0.4 \mu\text{g/m}^3$ NCC in TSP (this study, Suva Point) relates to the $PM_{2.5}$ aerosol fraction.

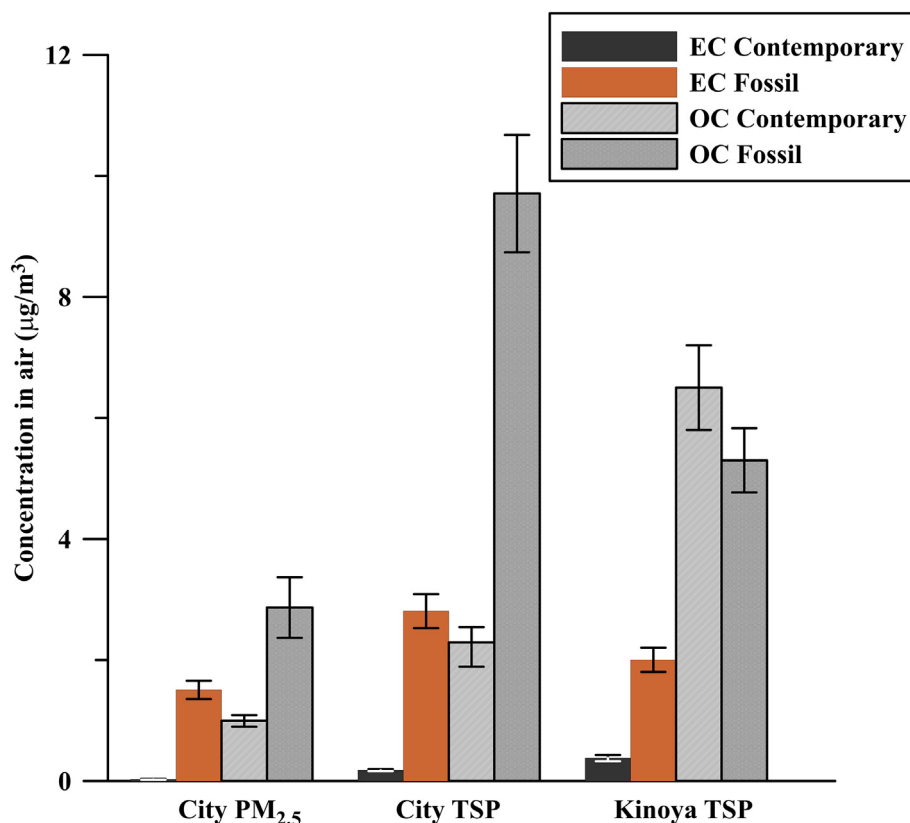


Fig. 3. Fossil and contemporary contributions to elemental and organic carbon particulate in Suva, Fiji, 2014–2015. Data for individual samples and notes on calculations and errors are included as Supplementary Table S4 (EC) and Table S5 (OC).

The ratio of OC/EC provides information about sources of carbon in Suva's air. Values for OC/EC for each site and for contemporary and fossil fractions are listed in Supplementary Table S6. Lewis et al. (2004) reports that OC/EC ratios larger than typical primary OC/EC ratio for an airshed ($(OC/EC)_p$) signal the onset of secondary organic aerosol. The $(OC/EC)_p$ was calculated for Suva for both $PM_{2.5}$ (1.69) and TSP (2.5) samples, by noting the minimum OC/EC ratio over all samples. Mean OC/EC for Suva was 2.5 (City $PM_{2.5}$); 4 (City TSP) and 5 (Kinoya TSP). These indicate the presence of secondary organic aerosol, from biogenic and fossil sources (Lewis et al., 2004). This OC/EC ratio changes for different assumptions about the OC/EC split, that is, when accounting for possible error in EC determination (Section 2.3.2). These changes were taken into account when solving Eq. (5) for sources (Section 3.3, Supplementary Table S7).

As for NCC, both EC and OC mass are greater in TSP than in the $PM_{2.5}$ fraction. For City samples, there is no clear difference between percentage contemporary NCC for $PM_{2.5}$ and TSP (compare (Masalaite et al., 2016)). As per south Asian studies (Gustafsson et al., 2009), the EC fraction was more fossil rich than OC. For fossil carbon, OC/EC ratios were all slightly lower than for NCC, ranging from 2 in City $PM_{2.5}$ to 3.5 in City TSP (2.6 in Kinoya TSP).

The largest fossil v. contemporary difference is in the OC fraction. This is also reflected in the OC/EC ratios for contemporary carbon which are >10 for each site (Supplementary Table S6). This indicates that biogenic secondary organic aerosols (Section 3.3.2) are a source of interest. As for NCC, at Suva City, OC was largely fossil carbon; whilst Kinoya OC had a larger proportion of contemporary carbon.

3.3. Sources

Estimated carbon contribution from different sources (Fig. 4) provides valuable information for source apportionment purposes. The uncertainties in the data used (Sections 2.4–2.5) have resulted in a wide range of potential values (error bars in Fig. 4 and details in Supplementary Table S7) for some sources, particularly OC from biomass and biogenic sources.

3.3.1. Elemental carbon

3.3.1.1. Fossil EC. Most EC in Suva City was from fossil sources. The EC/TC ratio may be used to infer some information about sources. For example, Schichtel et al. (2008) report EC/TC ratios characteristic of diesel vehicle emissions to be 0.61 (range 0.01 to 0.94) and 0.34 (0.01 to 0.91) for light duty gas vehicles. For Suva (Supplementary Table S6), mean EC/TC was 0.20 for total NCC or 0.28 if considering only $PM_{2.5}$. In comparison with data from Schichtel et al. (2008), this could indicate significant contributions from light duty gas vehicles, however the ranges reported (Schichtel et al., 2008) are too broad to draw any firm conclusions about diesel or gasoline source contributions.

It is known from emissions inventory studies that Suva's fossil fuel combustion sources in the city area include traffic (Isley et al., 2016). Diesel is a common automotive fuel in Fiji (Fiji Bureau of Statistics, 2014); as well as being used for shipping, industry and electricity generation (McDowall, 2005; Isley et al., 2016; Isley et al., 2018b). The use of low-grade diesel fuels in road vehicles, with up to 500 ppm sulphur content for Fiji (United Nations, 2014) increases $PM_{2.5}$ emissions by around

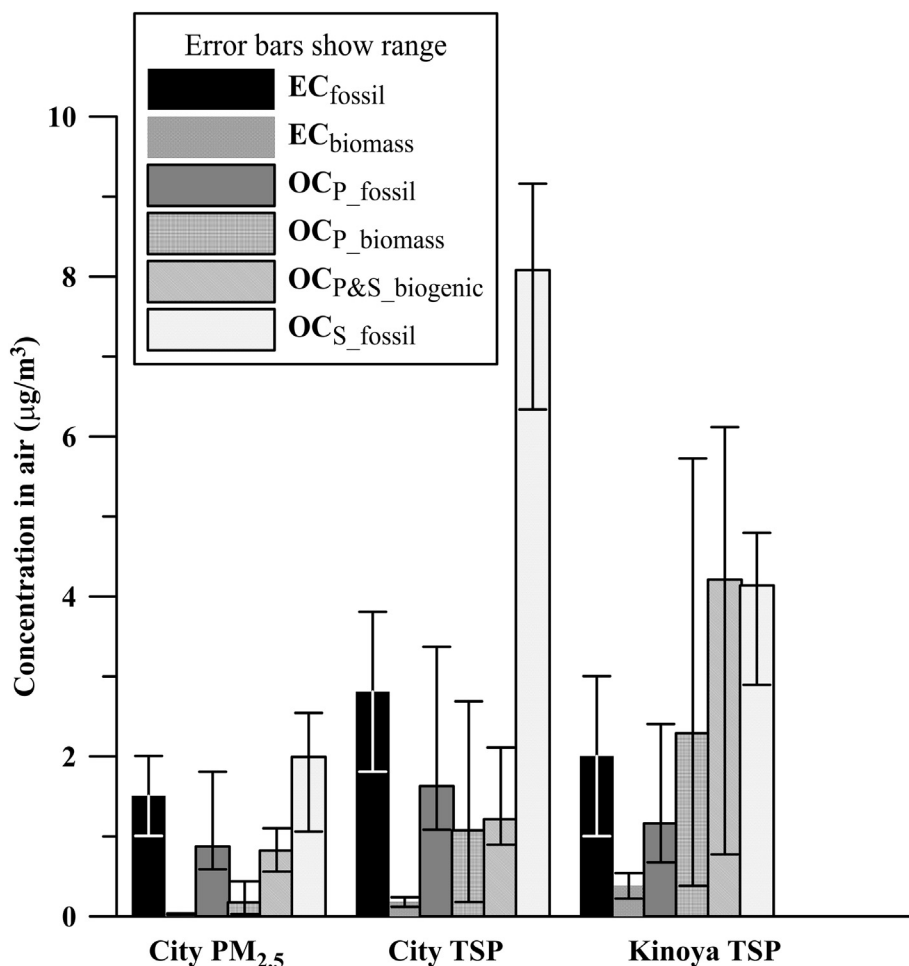


Fig. 4. Sources of carbon in Suva's aerosol. Data are included as Supplementary Table S7. Note that secondary organic carbon from biomass burning sources may also contribute to the calculated value of $OC_{biogenic}$.

30% (Boulter and Latham, 2009) in comparison to fuels with 10 ppm sulphur content (Australian Government, 2008). Similarly, waste oil usage by industries in Suva (McDowall, 2005) significantly increases PM_{2.5} emissions (3.26 ± 0.72 t/ML) in comparison with industrial diesel (25.1 ± 5.0 kg/ML) (Australian Government, 2011). Two power generation plants, burning diesel and heavy fuel oil (fossil sources), are also located at Kinoya (Keruring van Elektrotechnische Materialen te Arnhem, 2012).

Suva's particulate sources (PM_{2.5}), determined for this same time period by positive matrix factorization of elemental concentrations (Isley et al., 2018b), showed that around half of Suva City's PM_{2.5} mass was from sources with a fossil fuel combustion component. These included (Isley et al., 2018b): fossil fuel smoke (21%, shipping and industry); road vehicles (17%); industry (4%, cement and quarrying) and heavy fuel oil (3%, large ships). This study confirmed that fossil fuel emissions contribute significantly to Suva's aerosol. Elemental fossil carbon accounted for approximately 28% of NCC mass in City PM_{2.5}, 18% and 14% of NCC in City and Kinoya TSP respectively. Further to this, organic fossil carbon components (Section 3.3.2) also contributed significantly to NCC mass (Fig. 4).

3.3.1.2. Contemporary EC. The contemporary EC component, representing biomass burning (EC_{biomass}), was the smallest of all the sources identified for each site. Carbon concentration due to EC_{biomass} was more than double at Kinoya compared to the city. This reflects activities such as the burning of household waste and greenwaste and of cooking with biomass materials that occur more prevalently in Suva's residential areas (McDowall, 2005).

3.3.2. Organic carbon

3.3.2.1. Fossil OC. Again, fossil sources of OC are more prevalent in samples from the city area than those from Kinoya. Fossil sources of organic combustion include primary products of fossil fuel combustion (OC_{P_fossil} in Fig. 4).

Calculated anthropogenic secondary organic aerosols (OC_{S_fossil}), formed in the atmosphere from primary fossil fuel emissions, are more than double OC_{P_fossil} concentrations at each location sampled. For the city area, OC_{S_fossil} is the largest of the sources identified. The higher OC_{S_fossil} at Suva City is likely due to the higher concentration of road traffic, shipping and industrial activity near Suva City in comparison to the residential area of Kinoya. Indeed, (Plaza et al., 2011) showed that secondary OC can increase quickly in urban areas during heavy traffic periods. At Kinoya, OC_{S_fossil} is also among the largest sources, contributing a similar carbon mass to biogenic sources.

The prevalence of secondary particles is perhaps surprising for Suva, which experiences windy conditions and low atmospheric residence times over the land area of the city (Isley et al., 2017a). Yet this supports Isley et al. (2018b), which showed $18 \pm 1\%$ of PM_{2.5} to be secondary particles, based on analysis of elemental concentrations.

3.3.2.2. Contemporary OC. Contemporary sources of OC include primary OC from biomass burning. For Suva, this includes waste (largely biomass) burning (Gelencsér et al., 2007; Górká et al., 2014; Isley et al., 2016) and cooking emissions (Zotter et al., 2014). For the city area, OC_{P_biomass} contributes <10% to total NCC mass; with 16% at Kinoya.

Biogenic aerosols contribute secondary organic aerosols formed from volatile organic compounds emitted by vegetation (Claeys et al., 2004; Gelencsér et al., 2007; Bosch et al., 2014). Other primary biological particles contributing to these Suva measurements would include pollen and soil (Heal, 2014) as well as wear from tyres composed of natural rubber (Glasius et al., 2011). Biogenic carbon was the largest of the carbon sources determined for Kinoya (Fig. 4). As previously mentioned, this calculated biogenic component is also likely to have captured contributions from secondary organic aerosol resulting from biomass burning.

The contemporary carbon result for Kinoya, with more contemporary EC and OC than the city, is important as it provides some explanation for the differences seen between the emissions inventory developed for Suva (Isley et al., 2016) and source apportionment studies (Isley et al., 2018b). The source apportionment study (Isley et al., 2018b) was focussed on the city area and showed waste burning to contribute a much smaller PM_{2.5} mass fraction (8%) than fossil fuel burning (38% inclusive of vehicle sources). Kinoya results (Fig. 3) support the emissions inventory data (Isley et al., 2016), showing that, in the residential area, the impact from activities such as burning of contemporary carbon materials (including biomass wastes) and cooking is significantly greater than for the city area. Although not included in the emissions inventory (Isley et al., 2016), Fig. 4 demonstrates that biogenic activity in the Kinoya area is also more significant to aerosol mass than in the city.

4. Conclusion

Differences in total NCC concentrations between sites show that land-based emission activities contribute significantly to carbon concentrations in air at Suva City and Kinoya residential area. Even given the limited number of samples analysed, the differences between city and residential areas are clear. Analysis of further samples would provide data with greater accuracy for source apportionment. It would also be of benefit to analyse samples from other locations across the Pacific Islands. Contemporary carbon percentages in Suva City samples indicate that total NCC in air is primarily fossil carbon, from vehicles, shipping and industry; whilst in the residential area, around half of the total NCC is contemporary carbon; reflecting the higher incidence of biomass and waste burning, cooking activities and biogenic emissions. These results support the source apportionment for Suva City PM_{2.5} (Isley et al., 2018b), yet show that in the residential area, biomass combustion and biogenic emissions contribute more PM_{2.5} mass than fossil fuel burning. Air quality management strategies should therefore target open burning activities as well as fossil fuel combustion.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.06.182>.

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Paper Six

Ambient air quality and indoor exposure: PM_{2.5} implications for health in Suva Fiji

Isley C. F., Nelson P. F., Taylor M. P., Morrison, A. L. Atanacio A. J., Stelcer E. and Cohen D. D. 2018. *Air Quality and Climate Change*. 52(1): 35-43.

Supplementary information in Appendix E

This paper explores the differences in airborne particle composition across Suva. Expanding on the differences in PM_{2.5} and carbon noted between city, residential and coastal areas in earlier papers, this study considers elemental concentrations of particulate in a range of indoor and outdoor environments.

This adds an important comparison to Paper Four, which explored health risk, based on results from one site in Suva city. It shows that, because air quality may vary significantly across a city area, the health risk also varies with this differing exposure.

This paper also considers indoor sources, showing that use of wood and kerosene for cooking and lighting influenced indoor air quality. Indoor air quality was also influenced by outdoor sources, such as proximity to roads and industrial areas.

Use of polycarbonate filters in Paper Six meant that organic content is not able to be accurately quantified from these samples. Instead, carbon content (elemental and organic), determined for samples on quartz filters in Paper Five, provides more detailed analysis of organic aerosols across Suva.

Ambient Air Quality And Indoor Exposure: PM_{2.5} Implications For Health In Suva Fiji

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ABSTRACT

Air quality data collected at urban background locations is often assumed to represent a wider urban area. Localised sources and conditions can however cause variation between different microenvironments in the same urban area. Differences in PM_{2.5} (particulate less than 2.5 µm) composition may also have greater implications for health outcomes than PM_{2.5} concentration considered alone. Samples of PM_{2.5} were collected for three outdoor and nine indoor microenvironments across Suva, Fiji in 2014/15. Elemental concentration data have been used to estimate source contributions to PM_{2.5} for each site. The 12 sites are compared to concurrent ambient measurements at a fixed monitoring site in Suva City and to ambient photometer data. The objective is to determine how well ambient measurements represent air quality across the city, including indoor environments. Surveys were used to determine how much time is spent indoors and outdoors by Suva residents to ascertain potential exposure risks. Results show that PM_{2.5} concentration and composition varies significantly between the different microenvironments studied. Indoor air quality was affected by both ambient air and indoor sources. Fuel used for cooking, particularly wood and kerosene, influenced indoor PM_{2.5} and black carbon. Given that the survey showed that people spend more time indoors than outdoors, as experienced elsewhere in the world, ambient measures of PM_{2.5} concentration and calculated related health risk does not accurately reflect exposures arising from city indoor microenvironments.

Abbreviations:

ANSTO	Australian Nuclear Science and Technology Organisation
ASP	Aerosol sampling program air sampler
BC	Black carbon
EC	Elemental carbon
OC	Organic carbon
PM _{2.5}	Particulate matter with effective aerodynamic diameter less than 2.5 µm

Keywords: indoor, ambient, microenvironment

INTRODUCTION

Many air pollution health risk assessments or epidemiological studies use ambient monitoring data as an exposure proxy for local inhabitants. This can lead to errors in estimating exposure (Baxter *et al.* 2017; Park and Kwan 2017). Outdoor and indoor air may contain significantly different concentrations of both total PM_{2.5} and individual elements, yet in some cases there is correlation between the indoor and outdoor concentration (Molnár *et al.* 2007). This may occur at night, when human activity is reduced (Long and Sarnat 2004), and at times of year when the indoor-outdoor air exchange rates are increased (e.g. summer months in Canada, Bari *et al.* 2015). Activities such as heating and air conditioning significantly affect the relationship between outdoor concentrations and indoor exposures (Meng *et al.* 2012). Relationships between indoor and outdoor concentrations vary considerably within and between homes (Kearney *et al.* 2014); dependant on air exchange rates, particle size distribution, housing design, meteorology and occupant behaviour (Bari *et al.* 2015). Kheirbek *et al.* (2013) have shown that spatial and temporal differences in PM_{2.5} concentration are related to disparities in health impact between different neighbourhoods of the same city. Personal exposure is even more complex, and is also not necessarily equal to ambient PM_{2.5} concentration and composition or even to that measured indoors (Rao *et al.* 2015; McGrath *et al.* 2017).

Aerosol particulates from both indoor and outdoor sources are important contributors to residential exposures. Indoor PM_{2.5} is influenced heavily by indoor activities (Isaxon *et al.* 2015), particularly combustion of solid fuels (Butt *et al.* 2016). Exposure is greater for households cooking with solid biomass fuels, such as cow dung, agricultural residue and firewood, than with liquefied petroleum gas (LPG) and depends on kitchen design (Sidhu *et al.* 2017). Ambient conditions, particularly BC concentration (Götschi *et al.* 2002; Cao *et al.* 2005) and proximity to major roads (Rodríguez *et al.* 2008; Pérez *et al.* 2010; Kwon *et al.* 2016) also influence indoor exposures (Martins and Carrilho da Graça 2017). Higher exposures to outdoor air occur in homes with higher air exchange rates due to open windows (Baxter *et al.* 2017). Varied emission rates from traffic and biomass burning sources, ventilation factors and distance from sources result in rapidly changing air quality (Rodríguez *et al.* 2008; Laumbach *et al.* 2010).

The source of indoor PM_{2.5} may be more important for health implications than PM_{2.5} concentration. Black carbon (Hoek *et al.* 2013) and sulphur (Chung *et al.* 2015) carry a higher risk than many other PM_{2.5} components. As a result, for Suva, reduced annual mortality estimates due to elimination of ambient PM_{2.5} from fossil fuel combustion (potential reduction of 59 deaths), road vehicle emissions (20 to 35 deaths) and waste burning (25 deaths) surpassed predictions based solely on the PM_{2.5} mass of these components (11, 9 and 4 deaths respectively (Isley *et al.* 2018)). In comparison to Fiji's national road toll of 41 deaths in 2012 (Fiji Land Transport Authority 2013), mortality risks from Suva's ambient air pollution are significant and a reduction of pollutant sources would likely result in health benefits. Globally, air pollution from biomass burning and traffic sources constitute a major preventable cause of increased incidence and exacerbation of respiratory diseases (Laumbach *et al.* 2010). Indoor biomass burning is associated with adverse health outcomes including lung cancer (Raspanti *et al.* 2016), respiratory infections (Sigsgaard *et al.* 2015), low birth weight (Epstein *et al.* 2013), cataracts, cardiovascular health and all-cause mortality in adults and children (Fullerton *et al.* 2008; Bruce *et al.* 2015). Kerosene use for cooking and lighting are also associated with adverse respiratory symptoms (Choi *et al.* 2015) and low birth weight (Epstein *et al.* 2013).

Whilst Suva City sources and health risk of ambient fine particulates have been well characterised (Isley *et al.* 2018), indoor sources of exposure remain unquantified. This study aims to address this knowledge gap by determining the composition of indoor air across Suva. These data will be used in order to ascertain if ambient air sampling at Suva City adequately describes the exposure and health risk experienced at indoor locations.

METHOD

Study design

This study compares gravimetric particulate (PM_{2.5}) data, collected from 12 monitoring locations across Suva (reported in this study) to previously reported gravimetric measurements for a central location in Suva City. The Suva City ASP samples (Section 2.4), detailed in previous studies (Isley *et al.* 2017a), formed the basis of health risk calculations for Suva (Isley *et al.* 2018). Comparison of the two data sets,

to determine implications for health risk, is summarised in Figure 1.

Additionally, ambient photometer measurements of $PM_{2.5}$ are also available for the same period at three locations (Isley *et al.* 2017a): Suva City, Kinoya (residential) and Suva Point (ocean influenced). These provide an additional line of evidence for the variation in air quality across the greater Suva area.

Locations

Particulate matter samples ($PM_{2.5}$) were collected at 12 monitoring locations across Suva over the period October 2014 to October 2015 (Figure 2; Table 1) using Ecotech Microvol air samplers. Visual imagery of sampling sites is provided in Supplementary Section B.

Outdoor sites

Outdoor Microvol samplers were located in the Port offices area of the city, the industrial area of Walu Bay and a mainly ocean-influenced site at Suva Point. To allow comparison with previous studies (Isley *et al.* 2017a; Isley *et al.* 2018), Microvol samplers at Suva City, Kinoya and Suva Point were located as close as possible to ambient particulate sampling at these sites. For security reasons, however, the Microvol sampler at Kinoya was placed indoors and at Suva Point the Microvol sampler was located on a semi-enclosed balcony.

Indoor sites

These locations represent a range of Suva neighbourhoods, plus a more rural location, Navua. Toorak and Flagstaff are inner-city neighbourhoods of Suva City Council; Caubati and Kinoya lie within Nasinu, Fiji's most densely populated area, containing a mixture of formal and informal (squatter) settlements. Namadi Heights is an elevated area with more costly housing and Wainbuku is an outer residential area along the busy transport corridor to Nasouri (airport and farming region). A questionnaire was used to gather information about whether windows were open, what fuel was used for cooking, whether anyone was smoking nearby and if there were any nearby fires or air pollution events during sampling.

Sampling

Samples were collected from each of the sites in Table 1 using a portable Ecotech Microvol sampler. Polycarbonate filters (47 mm diameter) were exposed for approximately 24 hours using an Ecotech Microvol fitted with a $PM_{2.5}$ cyclone with flowrate of 3 L/min (as per Australian air sampling standards AS/NZS 3580.9.9 2006 and AS/NZS 3580.9.10 2006). Samples at Navua were each exposed for three days due to access limitations.

Participants for all locations reported that no smoking occurred during sampling. Interestingly, most participants reported there were no fires nearby during sampling, which was contrary to the frequent observation of small waste-burning fires at the time of sample collection. Domestic waste-burning is routine practice in Suva (McDowall 2005; Department of Environment Fiji 2007; Isley *et al.* 2016). Use of cooking fuels was also

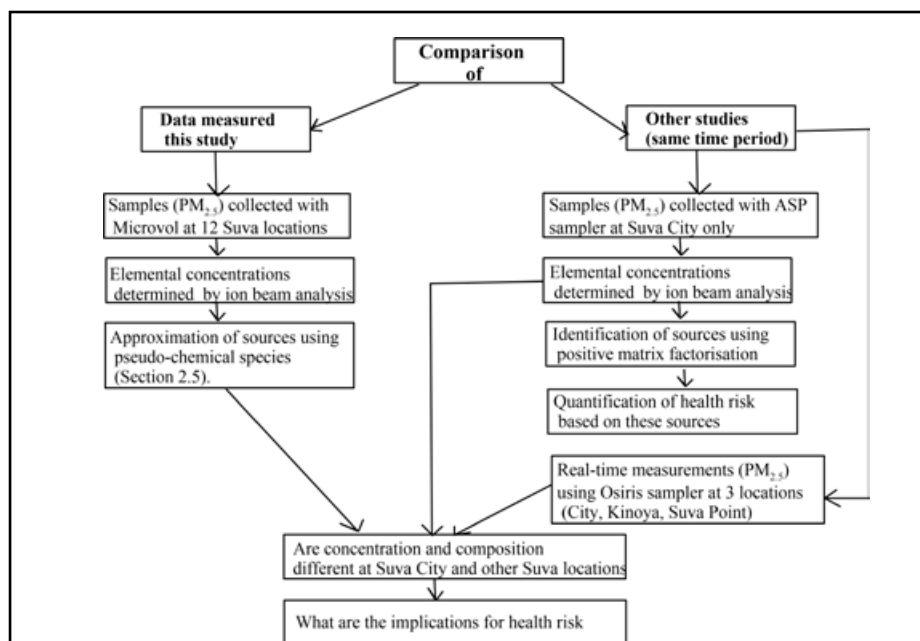


Figure 1: Study design. Data measured this study is compared against data from other studies for which sampling was completed during the same period (2014–2015).

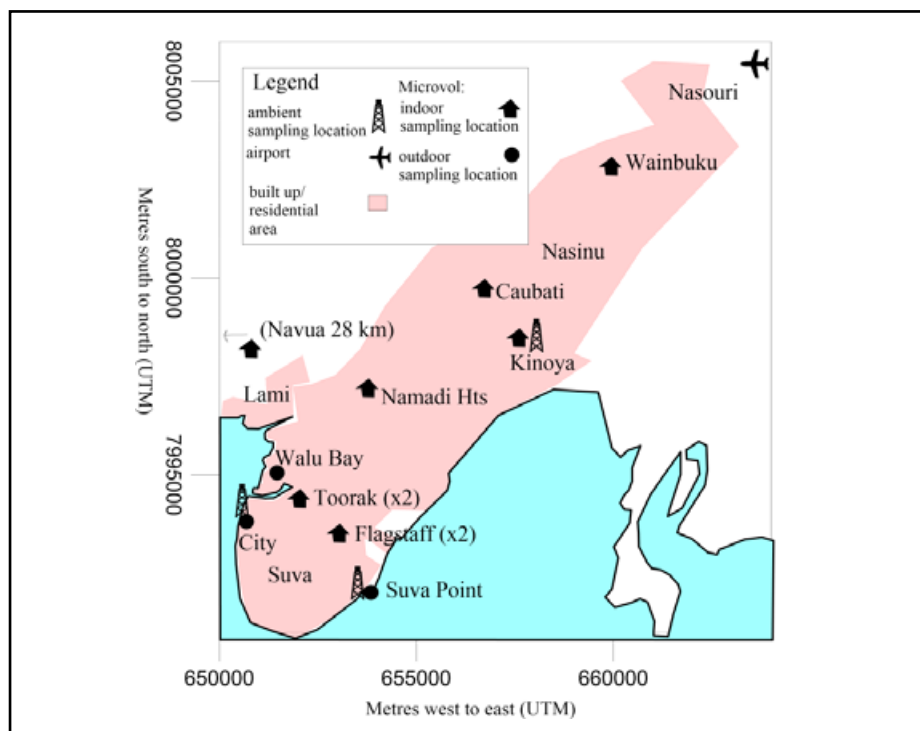


Figure 2. Sampling locations for ambient and indoor sampling in Suva, Fiji.

recorded. Namadi Heights reported a large number of households setting off fireworks (Diwali 22 October 2014). Occasional fireworks were also reported at Caubati.

Concurrent data for comparison

During the same period, ambient $PM_{2.5}$ samples were collected on 3 μm pore size stretched Teflon filters from Suva City using an Australian Nuclear Science and Technology Organisation (ANSTO) Aerosol Sampling Program (ASP) sampler (Isley *et al.* 2018). Additional $PM_{2.5}$ data, collected using Turnkey Osiris samplers was available for Suva City, Kinoya and Suva Point (Isley *et al.* 2017a). Relevant data from these concurrent studies that relate directly to

sampling dates of Microvol samples, are included in Supplementary Table S1, and discussed throughout this paper. Sampling and laboratory procedures for ASP and Osiris are presented in Isley *et al.* (2017a), along with results. Elemental concentration from ASP samples was determined by ion beam analysis (Isley *et al.* 2018).

Laboratory analyses

Gravimetric mass, black carbon concentration and elemental mass were determined on Microvol samples at the ANSTO laboratories, Sydney, Australia. The methods of analysis for Microvol samples, summarised below, were identical to those used on Suva City ASP $PM_{2.5}$ samples (see Isley *et al.* (2018)).

Site	Represents	Description	Windows open (% of time)	Cooking fuel	Height in m (Elevation)	Number of samples	Date sampled
<i>Indoor locations</i>							
Toorak 1 Brown St	Inner city living	Apartment, living area.4 occupants, 500m from Walu Bay industry.	100*	LPG	5 (39)	5	17-22/10/14
Toorak 2 Anand St	Inner city living	Cottage, kitchen.1 occupant, Supermarket 100 m away burns rubbish 3 times a week.	100*	LPG	2 (32)	10	20-30/4/15, 12-13/5/15
Caubati Matua Rd	Heavily populated residential area	Barracks (terraced units), Centre of house (living room); 3 occupants. Near informal settlements. Waste burning & occasional fireworks nearby	100	LPG	2 (45)	6	11-17/10/14
Kinoya near Kings Rd	Heavily populated residential area	Preschool, hallway.20 children, 1 resident. Power station (Diesel & residual oil) 1.3 km SE. Waste burning noted nearby.	100*	LPG	2 (33)	6	28-31/10/14 24-27/4/15
Namadi Heights Vunakece Rd	Wealthy area	House, 4 occupants. Enclosed veranda, Diwali fireworks Oct 22 nd 2014.	100*	LPG	2 (67)	10	9-31/10/14
Navua 28 km SW of Suva	Rural, wood & kerosene cooking	House, kitchen, walls black with soot. Adjacent room has wood-burning stove. 2 occupants, Close to ocean. Rice and cattle farming area	50	Wood (main) Kerosene	2 (7)	3 (for 3 days each)	3-12/5/15
Wainbuku Kings Rd 13 km NE of Suva	Northern suburbs	House, living room. 3 occupants. 70 m from busy road	100*	LPG	2 (24)	8	24-31/10/14 22/10/15
Flagstaff 1 Laucala Bay Rd	Workplace	Office waiting room, 10 workers, 1 resident, near main road	50	NA	2 (40)	5	16-22/10/14
Flagstaff 2 Bau St	City living	Apartment, 1 occupant Living room Faces away from street. Windows open 50% of time.	70	LPG	12 (52)	8	20-28/10/15
<i>Outdoor locations</i>							
City Kings Wharf	City	Roof of office building, port area, 130 m from bus terminal	NA	NA	17 (20)	4	21-25/4/15
Suva Point USP lower campus	Background	Balcony, semi-enclosed, ocean exposure. University campus area, on jetty	NA	NA	7 (9)	7	24-28/10/14 23-24/4/15 11-12/5/15
Walu Bay Foster Rd	Industrial area	Roof of office building, Business 100 m away burns cardboard; Near steel mill that burns waste oil	NA	NA	12 (21)	5	12-15/5/15

Table 1. Characteristics of Microvol PM_{2.5} sampling locations, Suva, Fiji. Participants reported that no smoking occurred at any of these sites during sampling. Locations marked with asterix (*) had windows that do not close, i.e. bars or mesh without glass.

Filter and exposed samples were weighed before and after exposure, at approximately 22°C and 50% relative humidity. Black carbon (BC) was measured on filters, before and after exposure, using the Laser Integrated Plate Method (Taha *et al.* 2007) at wavelength 633 nm and a mass absorption coefficient value of 7 m²/g. These PM_{2.5} mass and BC measurement methods are comparable to Beta Attenuation Monitors, IMPROVE-A thermal optical methods and thermal desorption methods (Hibberd *et al.* 2013; Hibberd *et al.* 2016). Elemental concentrations were determined by ion beam analysis on a 2 MV Tandemron accelerator. Three simultaneous techniques were applied for polycarbonate filters: particle-induced X-ray emission for concentrations of elements from Al to Pb; particle-induced gamma-ray emission for Na, F, Al, Mg and Li and Rutherford backscattering to determine total C, N and O. Hydrogen was unable to be determined for samples on polycarbonate filters. PIXAN analysis code was used to

extract the elemental concentrations from the spectrum peaks, as per Cohen *et al.* (2014). The average detection limit for the particulate matter species on polycarbonate filters was 0.02 µg/cm², ranging from 0.006 µg/cm² for Ni to 0.23 µg/cm² for Sr. One field blank was collected for every ten exposed filters.

Source contributions

Source contributions have previously been determined for Suva City ASP samples, using positive matrix factorisation (Supplementary Figure S1, (Isley *et al.* 2018)). Microvol sample numbers for each site were insufficient to perform multivariate statistical analysis of sources. Instead, source contributions to Microvol samples were estimated by using elemental concentrations to calculate a range of pseudo-chemical species. The equations below, developed by Malm *et al.* (1994) as part of the IMPROVE program in the United States, allow aerosol species to be estimated from the ratio of elements predominantly associated with each source and in this case

provides a means of basic PM_{2.5} composition comparison between sites. This method has been used previously for studies of PM_{2.5} sources across the Asia-Pacific region (Cohen 2010):

$$\text{Sea salt (sodium chloride)} = 2.54[\text{Na}] \quad (1)$$

$$\text{Ammonium sulphate} = 4.125[\text{S}] \quad (2)$$

$$\text{Soil} = 2.20[\text{Al}] + 2.49[\text{Si}] + 1.63[\text{Ca}] + 1.94[\text{Ti}] + 2.42[\text{Fe}] \quad (3)$$

$$\text{Biomass smoke} = [\text{K}] - 0.6 [\text{Fe}] \quad (4)$$

Limitations of the source approximation

This method provides an approximation of sources. Because the equations are based on broad source categories and not on chemical source profiles developed specifically for each site, it over-simplifies sources in comparison to techniques such as positive matrix factorisation. Even so, it provides a fairly simple way of observing basic differences in PM_{2.5} composition between the sites studied. Equation 1 assumes that sodium is

primarily associated with sea salt. This is supported for Suva (Isley *et al.* 2018) as Cl:Na ratios (Cl/Na = 1.56) represent fresh sea salt, with minimal Cl loss. Isley *et al.* (2018) does however also show association of Na with sulphates. Equation 2 assumes that sulphate is fully neutralised and occurs on the filters as ammonium sulphate. Although Isley *et al.* (2018) identified that most sulphur in Suva City's (ASP samples) $PM_{2.5}$ was associated with secondary sulphates (56%); other significant sulphur sources included road vehicles (20%) and smoke from open burning (15%). Soil (Equation 3) has been estimated from the oxide form of Al, Si, Ca, Ti and Fe. Given that H was not analysed on polycarbonate filters, the organic matter component was not determined. Traces included V, Cr, Mn, Co, Ni, Cu, Zn, Se, Br, Sr and Pb, which would, in practice, be attributable to various emission sources.

Reconstructed mass may be calculated (Cohen 2010) by adding Salt + Sulphate + Soil + Smoke + Traces + Organics + Black Carbon. The reconstructed mass is anticipated to be less than the gravimetric mass due to the absence of organics calculations, as well as the absence of nitrates and water vapour which are not measured with IBA. A significant fraction of nitrates can volatilise from the filters during collection and therefore may not be included in the gravimetric mass (Malm *et al.* 1994).

Organic content

Data addressing organic content from Suva are available in (Isley *et al.* 2017c). Whilst these are not directly comparable to the Microvol data, due to different sampling dates and fractions sampled, they provide relevant information about atmospheric organic content. Organic carbon (OC) in $PM_{2.5}$ was, on average, $3.8 \pm 0.6 \mu\text{g}/\text{m}^3$ at Suva City. This represents an average ratio of organic carbon (OC)/elemental carbon (EC) of 2.7 (range 1.6 – 4). For total particulate, OC concentration, at both Kinoya and Suva City was $12 \pm 1 \mu\text{g}/\text{m}^3$; accounting for one-third to one half of the average particulate mass (Isley *et al.* 2017a) in each case. These data confirm that there was significant organic carbon content in Suva's aerosol particulate during the 2014/15 sampling period.

By comparison, the undetermined content of the Microvol samples is on average 35% of total $PM_{2.5}$ mass. If this undetermined portion is assumed as organic content, it indicates a potential average OC/black carbon in $PM_{2.5}$ of 2.5 (over all sites, range 0.8 – 7). With BC approximately comparable to EC in other studies (Suva (Isley *et al.* 2017c), elsewhere (United States Environmental Protection Agency 2012)) it seems reasonable to assume that this unidentified content largely reflects organic matter.

Household surveys

In order to help compare Microvol data to typical exposures in Suva, household survey data were collected. A survey of 125 Suva households was conducted in October 2014. This included the occupants of indoor sampling locations used in this study, with the exception of Navua. This survey, intended

to characterise emissions and exposure, is described in further detail in Isley *et al.* (2016). Questions relevant to this study relate to the time spent indoors and outdoors; at work and at home. These questions were:

1. *Where do you spend most of your time during working hours?*
2. *How long do you spend at your place of work or study each day?*
3. *How much time would you spend indoors at home on a normal work day (include sleeping time)?*

Further detail on the survey questions and responses is provided in Supplementary Section C.

RESULTS AND DISCUSSION

$PM_{2.5}$ and BC concentrations

Overview of Microvol sampling results

Microvol sampling results, collected during short-term campaigns, provide a snapshot of $PM_{2.5}$ concentrations at that time. The short-term data do however provide a means by which ambient results at Suva City (ASP samples) may be compared against other locations. $PM_{2.5}$ and BC concentrations for each location are shown in Figure 3, with the data table included in Supplementary Table S1. Indoor concentrations may be anticipated to change with ambient conditions, activities and seasons (Section 1). Variation from one day to the next was significant at most locations (see Figure 3 for standard errors; cf. Ramachandran *et al.* (2000)). However, average $PM_{2.5}$ and BC concentration results from the two Flagstaff sites and two Toorak sites, which were sampled on different dates, show consistency (Figure 3). Samples from Suva Point, Kinoya and Wainbuku also represent a range of dates for each location (Table 1).

The spatial distribution of Microvol $PM_{2.5}$ and BC concentrations is in line with previous studies. That is, continuous Osiris $PM_{2.5}$ sampling data for 2014–2015 (Isley *et al.* 2017a), like the Microvol $PM_{2.5}$ data, show that $PM_{2.5}$ mass concentration at Suva Point is low, higher at Suva City and highest in the residential area of Kinoya (see also high volume air sampling data (Garimella and Deo 2007b)). Ultrafine particle number concentrations, another indicator of combustion aerosols, parallel the BC concentrations reported here, being low at Suva Point, moderate at Kinoya and higher at Flagstaff and Suva City (Isley *et al.* 2017b).

Comparison of co-located instruments

Microvol samples at the Suva City site showed agreement (within standard error for both $PM_{2.5}$ and BC, Figure 3) with concurrent ASP samples from the same location. The Osiris sampler, also located at this site, was not operational during Suva City Microvol sampling. Longer term results (one year 2014–2015, (Isley *et al.* 2017b)) show reasonable agreement (typically within 12%) between Osiris and ASP $PM_{2.5}$ at this City site.

Comparison of different locations

$PM_{2.5}$ concentrations measured in Suva City (ASP data) are different to $PM_{2.5}$ measured at the other sites (Figure 3). Pairs of concurrent $PM_{2.5}$ from Suva City (ASP) and each site (Microvol, including both indoor and outdoor sites) are not correlated $r = 0.05$, $p > 0.05$; for black carbon, $r = 0.17$, $p > 0.05$. The same holds when considering only indoor Microvol locations v ASP data or only outdoor Microvol locations v ASP data. Ambient $PM_{2.5}$ and black carbon concentrations for Suva City (ASP) do not predict accurately equivalent values for other Suva locations.

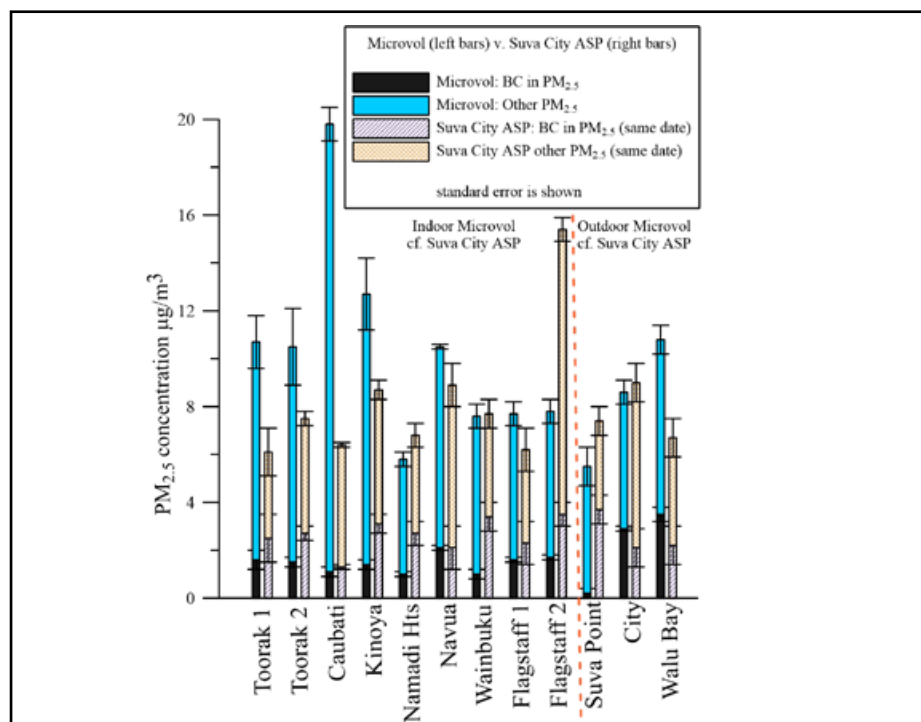


Figure 3. Comparison of Microvol $PM_{2.5}$ data at various sites around Suva to concurrent Suva City ASP $PM_{2.5}$ data. City, Suva Point and Walu Bay are outdoor sites, with the remainder being indoors.

Variation between Microvol sampling sites

Indoor Microvol $PM_{2.5}$ concentrations were lowest at Namadi Heights, at only 53% (average) of other indoor locations sampled. This is an elevated location, less likely to be affected by pollutants from surrounding areas and major roads. Caubati and Kinoya reported the highest indoor $PM_{2.5}$ concentrations. Waste burning would contribute to $PM_{2.5}$, with some settlements near these locations lacking waste collection services. Outdoors, the mainly ocean-influenced Suva Point location reported significantly lower $PM_{2.5}$ and BC than Suva City and Walu Bay (industrial).

Osiris data

Comparison within the same location

At Kinoya, the Microvol sampler, located indoors, reported $PM_{2.5}$ concentrations 20% higher than ambient (outdoor) Kinoya Osiris levels (Supplementary Table 1), for the same period. At Suva Point, the Microvol sampler, on a semi-enclosed balcony reported $PM_{2.5}$ levels 30% lower than those reported by the Osiris (ambient, Supplementary Table 1) at Suva Point for the same period. This indicates that indoor or even semi-enclosed environments may experience different air quality compared to nearby ambient conditions.

Comparison with other locations

Supplementary Table 1 shows data for Microvol $PM_{2.5}$ compared to the nearest ambient Osiris $PM_{2.5}$. Pairs of concurrent $PM_{2.5}$ from Osiris data and each site (Microvol, indoor and outdoor sites, excluding Suva Point and Kinoya co-located sites) are not correlated ($r = 0.3$, $p > 0.05$). The same is true when considering indoor and outdoor sites separately. Mean gravimetric $PM_{2.5}$ concentration for Microvol samples over all indoor sites ($10.3 \pm 1.4 \mu g/m^3$) were similar to longer term $PM_{2.5}$ averages from continuous ambient monitoring (Osiris) for Suva City and Kinoya ($9.7 \pm 0.1 \mu g/m^3$, 11.8 ± 0.4 respectively (Isley *et al.* 2017a)).

Elemental concentrations

Elemental composition of $PM_{2.5}$ varied by location. Average elemental concentrations over all Microvol sites ranged from $0.8 \text{ ng}/m^3$ for Co (Figure S2) to $1.3 \mu g/m^3$ for Cl. Concentrations reported for P, Cu, S, Zn, K, Ca, Br, Sr, V, Pb and BC were consistent with concentration ranges experienced in Suva City ASP samples (2014 to 2015, Supplementary Figure S2). Microvol $PM_{2.5}$ concentrations of Na, Al, Fe, Co, Si, Ni, Cl, Se, Ti, Cr, Mn were outside of this range for some locations, which are most likely due to localised sources.

Elemental concentrations in Microvol samples from Suva City are significantly higher than reported for the ASP data for Co, Cr, Mn, Se and Ti (Supplementary Figure S2). Different $PM_{2.5}$ cyclones have slightly different cut-points and sampling efficiencies (Turner *et al.* 2000; Cauda *et al.* 2014), which may explain differences in sampling results. The elements Co, Cr, Mn, Se and Ti are typically associated with soil and road vehicle wear/dust in Suva (Isley *et al.* 2018) and elsewhere:

(Lin *et al.* 2015; Peikertova and Filip 2015; Shirmohammadi *et al.* 2016), hence particle diameters are likely to be in the upper size-range of $PM_{2.5}$ (Taiwo *et al.* 2014; Gao *et al.* 2016; Pant *et al.* 2017). Suva Market is less than 0.2 km from this Suva City site and Garimella and Deo (2007b) reported total suspended particulate concentrations of Cr and Co at the Suva Market to be more than 150 times higher than the values reported here for the Suva City Microvol $PM_{2.5}$ samples. The Suva Market results reported by Garimella and Deo (2007b) are higher because they relate to a total suspended particulates, but demonstrate that these elements are characteristic of particulate in Suva City. Measurements of Mn, Se and Ti were not available for Suva Market in Garimella and Deo (2007b).

Sources of $PM_{2.5}$

Relative contributions of $PM_{2.5}$ sources varied by location (Figure 4), both for indoor and outdoor locations.

Salt

Sea salt accounted for 10% to 44% of Microvol $PM_{2.5}$ gravimetric mass at indoor locations and 8% to 31% outdoors. The data show that salt concentrations were highest at Navua corresponding to local coastal sources generated from more vigorous wave activity than present at Suva City (Wisuki 2017).

Further, the values recorded are within typical coastal values (Daniels 1989; Grythe *et al.* 2014). Salt variation between locations is typical of the variation of this source over time in Suva City ASP samples (Isley *et al.* 2018).

Sulphur

Navua (indoors) had higher sulphur concentrations ($0.40 \mu g/m^3$) than all other Microvol locations, both indoor and outdoor (Supplementary Figure S2). High sulphur concentrations at are likely due to cooking with wood and kerosene. In a review of kerosene emissions, Lam *et al.* (2012) note that sulphur emissions are not greatly influenced by device type but are primarily dependant on fuel sulphur content. Kerosene typically contains $<0.1\%$ sulphur (Lam *et al.* 2012). Firewood, the main cooking fuel used at this Navua location, typically contains around 0.13% sulphur (average of dry mass, Italy (Ozgen *et al.* 2014) c.f. (Jenkins *et al.* 1998) for different wood types). 'Yard waste' (Jenkins *et al.* 1998) contains around 0.24% sulphur (average of dry mass); hence nearby agricultural and waste burning may also contribute to the elevated values identified here.

Outdoors, Suva Point also reported a high proportion of sulphates (18 %, Figure 4), indicating potential contribution from biogenic ocean sources (cf. Isley *et al.* 2018).

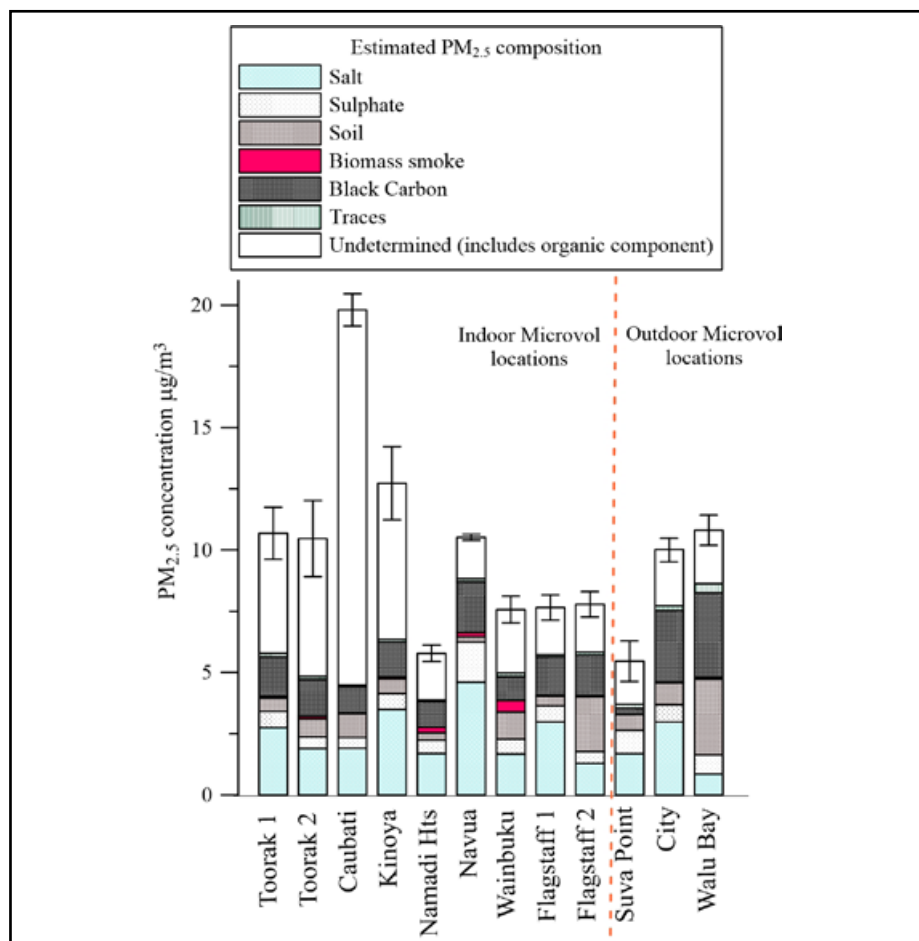


Figure 4: Gravimetric mass and estimated composition of Microvol $PM_{2.5}$ samples. Standard error of total gravimetric mass is shown. Bar charts show reconstituted mass of $PM_{2.5}$ constituents, determined from elemental concentration (calculated using methods in Cohen (2010)). As H was not analysed, the organic component was unable to be determined but is likely to form much of the difference between reconstituted and gravimetric mass.

Site	Microvol PM _{2.5}	Microvol BC	Distance from ambient monitoring site	Nearest ambient PM2.5 measurement (Photometer)	Gravimetric PM2.5-City (ASP)	ASP BC
<i>Indoor locations</i>						
Toorak 1	10.7 ± 1.1	1.6 ± 0.4	850 m W City	8.1 ± 0.1	6.1 ± 0.2	2.5 ± 1.0
Toorak 2	10.5 ± 1.6	1.5 ± 0.2	600 m W City	3.5 ± 0.1	7.5 ± 0.7	2.7 ± 0.3
Caubati	19.8 ± 0.7	1.1 ± 0.2	550 m NW Kinoya	8.3 ± 0.2	6.4 ± 0.2	1.3 ± 0.1
Kinoya	12.7 ± 1.5	1.4 ± 0.2	30 m Kinoya Indoors	10.4 ± 0.2	8.7 ± 0.7	3.1 ± 0.4
Namadi Heights	5.8 ± 0.3	1.0 ± 0.1	2.8 km SW Kinoya 3.5 km NE City	12.7 ± 0.2 9.1 ± 0.1	6.8 ± 0.5	2.7 ± 0.5
Navua	10.5 ± 0.1	2.1 ± 0.1	28 km SW City	10.6 ± 0.1	8.9 ± 2.2	2.1 ± 0.9
Wainbuku	7.6 ± 0.5	1.0 ± 0.2	7.2 km NE Kinoya	13.2 ± 0.3	7.7 ± 0.9	3.4 ± 0.6
Flagstaff 1	7.7 ± 0.5	1.6 ± 0.1	1.8 km SE City	8.3 ± 0.1	6.2 ± 0.2	2.3 ± 0.9
Flagstaff 2	7.8 ± 0.5	1.7 ± 0.1	1.7 km SE City	10.1 ± 0.1	15.4 ± 2.4	3.5 ± 0.5
<i>Outdoor locations</i>						
Suva Point	5.5 ± 0.8	0.2 ± 0.2	5 m Suva Point	8.3 ± 0.2	7.4 ± 0.8	3.7 ± 0.6
City	8.6 ± 0.5	2.9 ± 0.1	10 m City	equipment malfunction	9.0 ± 0.9	2.1 ± 0.8
Walu Bay	10.8 ± 0.6	3.5 ± 0.3	1.2 km NE City	equipment malfunction	6.7 ± 0.7	2.2 ± 0.8

Table S1: PM_{2.5} and BC concentrations for Microvol sampling locations, Suva, Fiji. Also shown are concurrent PM_{2.5} results from the nearest ambient monitoring site and concurrent gravimetric PM_{2.5} results from Suva City.

Whilst there are not shipping ports at Suva Point or Navua, off-shore shipping sources may also contribute.

Black carbon

Outdoors, BC concentrations were highest in the industrial area of Walu Bay, followed closely by Suva City (Figure 4); with BC concentrations at Suva Point being more than 10 times lower.

Indoor BC concentrations were highest at Navua, where wood and kerosene are used for cooking; these cooking practices are reflected in 17% and 20% (respectively) of Suva households (Isley *et al.* 2016). Wood burning stoves emit significant levels of carbon comprising 20%-60% of PM_{2.5} emissions (largely BC) for hot burning stoves (Rau 1989). Kerosene burning also emits BC at around 8% of PM_{2.5} emission (Reddy and Venkataraman 2002). Households using LPG for cooking appear to be more influenced by outdoor sources. Higher indoor BC concentrations were recorded in areas of increased vehicle traffic (Flagstaff) and industry (Toorak). Relevantly, Isley *et al.* (2017b) report ultrafine particle number concentrations to also vary considerably by location in Suva and to be significantly affected by transport emissions and proximity to roads. Homes in Fiji are typically constructed to allow airflow and may not have sealable windows, allowing continuous air exchange between indoors and outdoors (see visual imagery in Supplementary Section B). This was the case in several locations sampled here (Table 1).

Biomass smoke

Biomass burning estimates (Figure 4) were highest for Wainbuku, Namadi Heights and Navua, (all indoor sites) with wood known to be used for cooking at the Navua site.

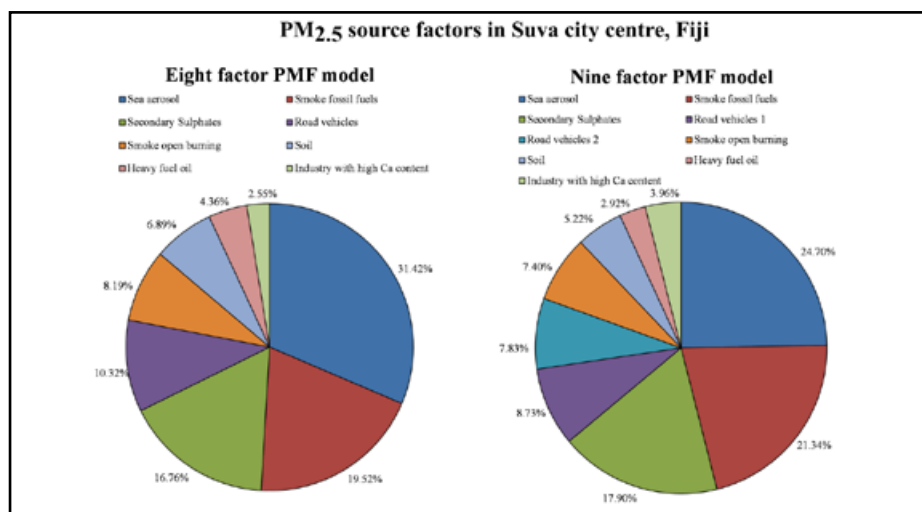


Figure S1: PM_{2.5} source factors for Suva City, 2014-2015. Data from Isley *et al.* (2018)

Estimated biomass smoke contributions (Figure 4) are lower than waste burning contributions reported for Suva City ASP samples (7% to 9% of PM_{2.5} mass (Isley *et al.* 2018)). Open burning emissions (household wastes and green wastes) would also contribute to the organic component of PM_{2.5} (Andreae and Merlet 2001; Schauer *et al.* 2001; Mohr *et al.* 2009; Akagi *et al.* 2011). In many parts of Fiji, agricultural burning from sugar cane farming would also contribute to biomass smoke, however sugar cane farming does not occur near Suva and therefore is not considered to be a factor for biomass estimates.

Soil

Outdoors, soil contribution was greatest at Walu Bay, potentially due to emissions from local industries, including vehicle movements on unpaved work-yards. The soil calculation includes Fe concentration and may be

overestimated due to Fe emissions from the nearby steel processing facility (Garimella and Deo 2007a). Relatively higher concentrations of Cr, Mn and Fe at Walu Bay may also show influence from local industries, particularly from the combustion of waste oil, which contributes to the higher S and BC at this location. Cement industry operations in nearby Lami may also contribute to Ca at Walu Bay.

Indoors, the two Flagstaff sites differ in terms of soil contribution to PM_{2.5}. The second Flagstaff site was sampled during a prolonged period of dry weather and windblown dust levels in the air were generally higher at this time (Isley *et al.* 2017b) also note higher Suva City ASP PM_{2.5} for this period (Figure 3)).

Undetermined portion of PM_{2.5}

On average, reconstituted mass, excluding

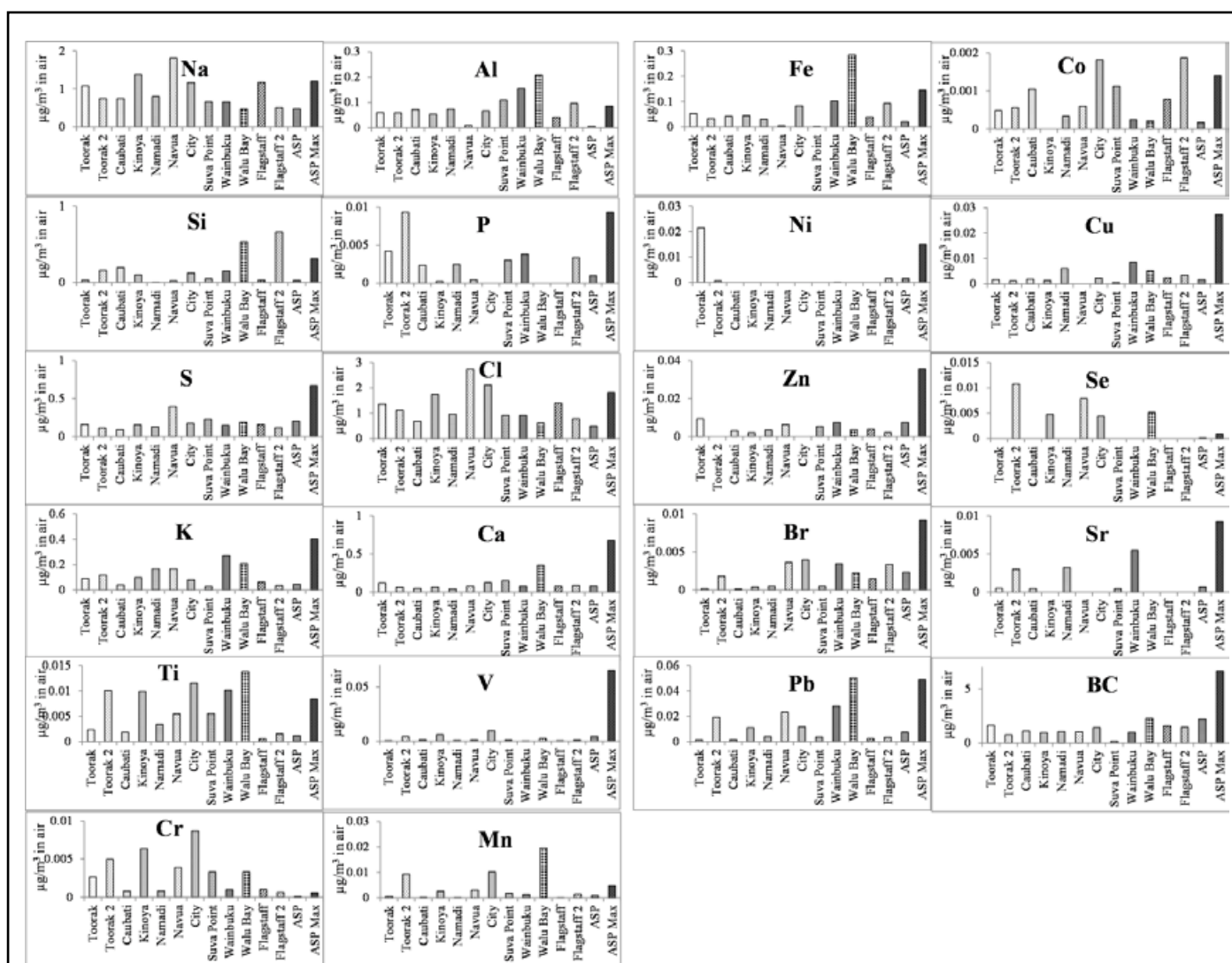


Figure S2: Elemental concentration data for each element, average over all Microvol samples for each site. ASP data included here is the annual mean of all samples collected at Suva City from October 2014 to October 2015. Maximum concentration recorded in ASP samples is also given in order to demonstrate the variation in elemental concentration recorded at the city site over a year.

organic components, accounted for 65% of gravimetric mass. This corresponds to the data from Cohen (2010), who observed that organic matter typically comprises up to 40% of $PM_{2.5}$ mass.

Time spent indoors and outdoors

Survey results (Supplementary Section C) indicate that respondents ($n=125$) spent around 16 hours per day indoors. This includes an average of 10.3 hours per day indoors at home. During working hours, 58% of respondents were inside an office, factory, classroom or house; 25% spent equal time working indoors and outdoors; 15% were outdoors and 2% in a vehicle. The average time spent indoors at work was therefore 5.6 hours. Data on employment in different professions was not available for the Suva area (Fiji Bureau of Statistics 2015) and the survey results may not accurately represent the number of outdoor workers. Notwithstanding this limitation, the survey data revealed that the surveyed Suva residents spent 67% of their time indoors. This is lower than reported in other studies, globally (Moschandreas 1981; Jenkins *et al.* 1992; Klepeis *et al.* 2001). Nevertheless, this time period is significant because it

indicates that Suva residents are exposed to indoor air for a greater period of time than outdoor air. Therefore, basing health risk assessment on outdoor air quality is likely to underestimate the airborne hazards and the consequent exposure and risk based on $PM_{2.5}$ concentration.

Health risk implications

Health risk data cannot be directly applied to the Microvol $PM_{2.5}$ concentration results. Firstly, these $PM_{2.5}$ results represent short-term air quality data and the health risk data (Hoek *et al.* 2013) are developed using long-term data. Similarly, risk data were developed using ambient concentration levels and the relationship to indoor levels has not been established. Notwithstanding this limitation, the variation of both $PM_{2.5}$ concentration and $PM_{2.5}$ composition observed here indicate that potential health implications may differ across the city. Health risk is typically assessed based on $PM_{2.5}$ mass (Hoek *et al.* 2013). Indoor locations had higher $PM_{2.5}$ concentrations (median $10.5 \mu\text{g}/\text{m}^3$ over all indoor locations) than concurrent ASP values (median $7.5 \mu\text{g}/\text{m}^3$). Thus, health risk based on Suva City ASP $PM_{2.5}$ (annual mean $7.4 \mu\text{g}/\text{m}^3$) values may therefore underestimate exposure risk to

particulates at these locations.

Health risk may also be calculated based on black carbon content (Hoek *et al.* 2013). For Suva City ASP samples, risk based on BC concentration (annual mean $2.2 \mu\text{g}/\text{m}^3$) exceeded risk based on $PM_{2.5}$ mass (Isley *et al.* 2018). Black carbon concentrations at all indoor Microvol sites (median $1.5 \mu\text{g}/\text{m}^3$) were lower than for Suva City ASP data (concurrent data median $2.7 \mu\text{g}/\text{m}^3$). Hence, use of Suva City ASP data may overestimate risk from BC at these locations. The same is true for risk based on sulphur content (Beelen *et al.* 2015), where only Navua (indoor) and Suva Point (outdoor) had higher sulphur concentration than Suva City ASP data. Basing health risk on one ambient monitoring location (Suva City) may not accurately represent risk at these different locations.

CONCLUSION

Concentration and composition of $PM_{2.5}$ varies considerably across Suva, both for indoor and outdoor locations. $PM_{2.5}$ concentrations were higher for indoor locations (compared to outdoors), whereas black carbon concentrations and sulphur were mostly lower indoors. Given that

both indoor and outdoor sources appear to influence indoor air quality in the locations sampled, ambient outdoor results are considered a poor predictor for indoor microenvironments and potential exposure risk. Given that Suva residents spend most of their time indoors (~16 hours per day), calculated health risk based on City ambient data may not accurately reflect *actual* health risks across Suva. These data indicate that indoor health risk, based on BC and S concentration, are greatest where indoor emissions from cooking contribute strongly to PM_{2.5}.

SUPPLEMENTARY SECTION

Supplementary data is available for this article. Section A includes figures and tables providing further data on sampling results in terms of PM_{2.5} mass, elemental concentrations and source contributions. Section B presents imagery of the sampling locations. Section C summarises the results of the household survey.

<https://drive.google.com/file/d/1aQGdiAsRV99jZ3yGJcLoOhG22SXfuXXw/view?usp=sharing>

ETHICS APPROVAL:

Official approval to conduct this research in Fiji was granted on the 28 April 2014, reference RA20/14 by Parmeshwar Mohan, Permanent Secretary for Education, National Heritage, Culture and Arts, Fiji and, on 12 March 2014, reference 12/2/1, by Eleni Tokadua, Ministry of Local Government, Urban Development, Housing and Environment, Fiji. Ethics approval was granted for this research by the Human Ethics Secretariat at Macquarie University (reference number 5201400557), dated 24 June 2014.

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Paper Seven

Air quality management in the Pacific Islands: A review of past performance and implications for future directions

Isley C. F. and Taylor M. P. 2018. *Environmental Science and Policy* 84: 26-33.

Supplementary information in Appendix F

This paper considers the sources of air pollution identified in Papers Three to Six in the context of environmental policy in the Pacific Islands.

The relevance of this paper to the overall thesis is that it considers the factors involved in formulating air quality goals and strategies that are relevant in the Pacific Island context. The focus of the paper encompasses not just Fiji, but all Pacific Island Countries and in doing so examines similar policy approaches across these small island nations.

More relevantly, this paper examines how the collected data may be applied practically, in order to bring about real improvement of environment and health to Pacific Islands nations.

Graphical abstract for Paper Seven





Review

Air quality management in the Pacific Islands: A review of past performance and implications for future directions

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ABSTRACT

Air quality is the leading global environmental risk factor for disease. This article focuses on the evidence for the need to develop effective air quality policies for the Pacific Islands region. Factors that have influenced the success and failures of previous and existing environmental policies are considered to help understand necessary future actions. Factors instrumental in resulting in policy failures include nations focusing on economic growth and poorly managing the externalities (i.e. waste and fossil fuel emissions); inappropriate application of aid; a lack of planning; insufficient resources; misunderstanding of risks and conflicts in systems of governance. Successful programs have included capacity building activities in collaboration with traditional land-users; empowering of existing leaders, regional co-operation and local acceptance of financial responsibility. Forward strategizing for more effective leadership in air quality management will require a more co-ordinated approach to address enforcement of environmental policy from multiple angles: including raising awareness, provision of viable alternatives, local financial responsibility and the co-operation of different authorities to facilitate enforcement.

1. Introduction

Globally, exposure to environmental pollution kills three times as many people each year than AIDS, tuberculosis and malaria combined; almost 15 times as many as war and all forms of violence (Landrigan et al., 2017). Air pollution is the leading global environmental risk factor for disease and premature death (World Health Organization, 2014). Risk from PM_{2.5} (particles less than 2.5 µm) includes all-cause mortality (Kloog et al., 2013; Shi et al., 2016), particularly cardiovascular mortality (Hoek et al., 2013) with no evidence of a threshold below which effects are not observed (Pope and Dockery, 2006; Brook et al., 2010). Other associations include diabetes (Potera, 2014; Rao et al., 2015); deep vein thrombosis and pulmonary embolism (Kloog et al., 2015), dementia, Alzheimer's and Parkinson's disease (Kioumourtzoglou et al., 2016). In Suva alone, the annual mortality risk from PM_{2.5} exceeds the national road toll for Fiji (Isley et al. (2018a)). The main sources of PM_{2.5} in Suva, Fiji's capital, are smoke from fossil fuel burning in industry, power generation, shipping and vehicles (Isley et al., 2016, 2017b, 2018a, 2018b) as well as smoke from burning wastes in residential areas (Isley et al., 2017a; Isley et al., 2018b).

The Fiji Department of Environment are aware of these air pollution sources (Department of Environment Fiji, 2007; Fiji Department of

Environment, 2013). Essentially, the same sources of air pollution (diesel combustion and waste burning) are common across all of the Pacific Island countries (PICs) and many other developing countries (Thaman et al., 2003; Periathamby et al., 2009; Gleye, 2010; Mataka, 2011; Owens et al., 2011; Dornan and Jotzo, 2012; Keruring van Elektrotechnische Materialen te Arnhem, 2012; Pacific Energy Summit, 2013; Wiedinmyer et al., 2014; Escoffier et al., 2016; Taibi et al., 2016; Isley et al., 2018a). The Pacific Island nations suffer from an absence of, or incomplete options for residential waste disposal, leading to individuals incinerating domestic waste. This causes localised air quality issues (Mataka, 2011; Woodruff, 2014a). Consequently, effective waste management forms an integral aspect of air quality policy (Department of Environment Fiji, 2007; Fiji Department of Environment, 2013) and is examined in this article as part of the matrix for lowering health risks from particulate pollution in Pacific Island cities.

For Suva, Fiji, scientific air quality studies are available to describe the current particle air quality and pollutant sources (Isley et al., 2016, 2017a, 2017b, 2018a). Similar information is largely absent across the PICs, with the exception of Noumea and New Caledonia (Gleye, 2013; Escoffier et al., 2016). For the city area in Suva, Fiji, diesel emissions from industry, electricity generation and small shipping craft contribute approximately 21% of the fine atmospheric particle (PM_{2.5}) load; with

Abbreviations: OECD, Organisation for Economic Co-operation and Development; PIC, Pacific Island Country; SPREP, Secretariat of the Pacific Regional Environment Programme

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17% from vehicle emissions, 7.5% from open burning and 7% from large ships and other industries. In the residential areas of Suva, the open burning contribution is more than double that in the city, largely due to waste burning and cooking practices (Isley et al., 2018b). In Noumea, industrial activity, traffic and domestic burning practices are notable sources of particle air pollutants, although quantitative data on their specific contribution is not available (Escoffier et al., 2016).

The article evaluates current air quality management in the Pacific Islands in the context of existing environmental management practices and known health risk. The aim is to determine the best practices for developing forward strategies for air quality management.

2. Existing policy

Much environmental policy exists across the PICs. This includes legislation (Fiji Environmental Law Association, 2017a; Secretariat of the Pacific Environment Programme, 2017). Participation of PIC's in 37 different international environmental treaties and regional frameworks and policies is listed in Secretariat of the Pacific Regional Environment Programme (2016). Air pollution is encompassed within this broader legislation, however, for Fiji, the Environment Management (EIA process) Regulations (Government of Fiji, 2007b) include a section on air emission licenses. The national ambient air quality standards are included within Environment Management (waste disposal and recycling) Regulations (Government of Fiji, 2007a); which include penalties for open burning of domestic waste, tyres, oil and other materials. Despite existing legislation, implementation and enforcement are seldom effective, both in Fiji (Fiji Environmental Law Association, 2017b) and elsewhere across the region (Papua New Guinea (Mowbray and Duguman, 2009), Micronesia (Government of the Federated States of Micronesia, 2007) and other Pacific Islands (Nunn, 2009).

For example, with respect to waste burning, Micronesia's law clearly states that the open burning of wastes is not allowed (United States Government 1980; Harding, 1992). Yet, *"the burning of yard waste, which may include plastics, rubber and many other inorganic items, is a common practice"* (Government of the Federated States of Micronesia, 2007). Waste burning continues in Micronesia despite advances in waste policy (Woodruff, 2014b). Fiji's open fires by-laws (Government of Fiji, 2007c) include a \$10,000 (FJD) penalty for burning household garbage without a permit. This penalty is severe, being half of the average annual wage in Fiji (Fiji Bureau of Statistics, 2015); yet there is no evidence of enforcement. Instead, surveys show that waste burning is widespread in Fiji with over 50% of people in Suva (capital) burning household waste and green waste (McDowall, 2005; Department of Environment Fiji, 2007; Isley et al., 2016). Imagery of waste-burning in Suva Fiji (Supplementary Section A), show that items burned are similar to those described for Micronesia. Likewise, for Kiribati, the Cook Islands and the Marshall Islands, enforcement of bylaws and policy around waste disposal is minimal and are ineffective at preventing burning of waste (Marshall Islands, 1984; Kiribati, 1999; Dusevic, 2001; Secretariat of the Pacific Environment Programme, 2013; Aitken, 2014).

Even where legislation is uplifted to remedy pollution caused by poorly maintained vehicles, past history has shown that it is difficult to implement. United States Government (1980) regulations prohibit use of vehicles in Micronesia that have become 'mechanically deficient so as to cause the emission of visible air contaminants' and list penalties. Smoky vehicles remain a problem despite these legal controls (Government of the Federated States of Micronesia, 2007; UNEP, 2015). Similar to Fiji's burning laws, this may be because the controls are *"unenforceably severe"* p.22 (Harding, 1992). The Fiji Department of Environment, (2013) listed vehicle emissions as the most common source of air pollution complaints. Since then, the number of registered vehicles has risen steadily, from 89,190 in 2013 (Fiji Bureau of Statistics, 2018); to 110,763 in 2016 (Vula, 2017). The most recently available data for the composition of Fiji's vehicle fleet is from 2013

(Fiji Bureau of Statistics, 2018), with 22% of vehicles being buses, carriers and goods vehicles. Whilst the Fiji Bureau of Statistics (2018) does not list data for the age of vehicles in Fiji's fleet, vehicle emissions from old and poorly maintained vehicles also remain an unresolved problem (Campbell, 2004; Rogo, 2011; Land Transport Authority Fiji, 2016). Fiji's Land Transit Authority Fiji (2015) notes that *"while it is true that all motor vehicles smoke, it is the degree of it which is the problem"* in Fiji (imagery in Supplementary Section B). Fiji's Land Transport Authority is currently taking action to reduce emissions, detailed in Section 4.3.

3. Barriers to implementing policy successfully

3.1. Economic barriers

Governments in PICs trend to focus on economic growth, with spending on environmental protection being a low priority (Nunn, 2009; Lata and Nunn, 2012; OECD, 2012). Growing expectations for development compete with management of impacts on a fragile environment (Storey and Hunter, 2010). This is driven by communities who are more focussed on short-term economic benefits (Lata and Nunn, 2012).

For those on low incomes, long term health risk or environmental goals seem less relevant than daily needs (Nunn, 2009). In the Federated States of Micronesia, 17% of the population lived below the \$1.90 per day income poverty line in 2013 (39% in Papua New Guinea (World Bank, 2017)). For many in informal settlements (squatters), day-to-day survival is the primary concern (Jones, 2013). A similar situation exists in Port Moresby (Papua New Guinea), where more than 375,000 live in informal settlements (50% of population (Jones, 2013)) and to varying degrees in Suva where 20% of the population are squatters (Phillips and Keen, 2016). Nadi (Fiji), Honiara (Solomon Islands), Port Vila (Vanuatu) and Apia, Samoa also have significant squatter populations (Jones, 2013). Issues such as climate change or long-term health impacts are a future problem, for which the timing and severity of the impact is uncertain. Understandably, these are often not given substantial consideration for those facing immediate daily challenges of poor housing, inadequate waste disposal, unemployment, nutrition-related health problems and under-resourced health services (Mortreux and Barnett, 2009).

Low income households are less likely to consider air pollution consequences when disposing of rubbish or choosing cooking fuel. Informal settlements lack solid waste collection services, because they do not pay land rates (Lal et al., 2007). In some instances, communities will pay for their own waste collection (Lal et al., 2007). Suva Council (Fiji) has installed bins at the edge of informal settlements to allow waste disposal (Phillips and Keen, 2016). Unfortunately, the frequency of waste collection is not always adequate (imagery Supplementary Section C). Burying wastes or burning them are often the only practical and hygienic options (Mataki, 2011). Therefore, policies prohibiting waste burning are unlikely to be effective, unless alternative options are available. Likewise, when choosing fuels for cooking, the poorest households are likely to choose high-emission fuels that can be freely collected, such as firewood and even plastics (Government of the Federated States of Micronesia, 2007), because cleaner fuels are priced beyond their reach (OECD, 2012).

This short-term perspective is also demonstrated in the transport sector, where motorists lack economic incentive to consider adverse environmental and health impacts, which are not borne by themselves, but by the general public (OECD, 2012). Transport is a vital component of Fiji's economy, contributing 16% to GDP (Fiji Bureau of Statistics, 2014). As diesel is considered to be Fiji's 'working fuel', it is taxed less in comparison to petrol (Rogo, 2011). This growth-oriented policy generates increased diesel emissions, competing with environmental policies that advocate emission reduction (Rogo, 2011). Further, lower taxes on diesel fuels are common across many countries, leading to an

increase in diesel vehicles (OECD, 2012).

Small countries cannot support large numbers of government employees and environmental departments that do not generate revenue are particularly understaffed (Nunn, 2009). This is common across developing countries (Momtaz and Kabir, 2014). Fijian municipalities receive inadequate funding to administer solid waste services (Fiji Department of Environment, 2010) and enforce waste burning policy (Government of Fiji, 2007c, 2012). Furthermore, there are insufficient (Government of Fiji, 2006) or inadequately trained (Tiake et al., 2002) inspectors to monitor offences. Collection of waste has high costs and is economically inefficient (Fiji Department of Environment, 2010). Programs that are economically driven often fail due to fuel cost, widespread populations and limited local markets for recycled goods (Kerr, 2005; Finnigan, 2011; Woodruf, 2014a).

3.2. Technology, infrastructure and planning

Inappropriate technology, lack of maintenance and a shortage of skilled workers often see new measures fail (Finnigan, 2011). Vehicles provided for solid waste collection may be inappropriate if they are unsuited to unpaved roads common in the informal settlements (Woodruf, 2014a). Maintenance of waste compactor vehicles and incinerators is difficult in PICs due to difficulty acquiring spare parts, equipment damage caused by hot, salty climate and shortage of skilled mechanics (World Health Organisation, 1996). Alternative solutions such as high cost solid waste landfills in the Cook Islands and Kiribati proved too complex in design (Finnigan, 2011), equipment failed and was not repaired due to a lack of skilled workers, spare parts or funds (Storey and Hunter, 2010).

International aid often addresses the need for environmental policy and programs in PICs (World Health Organisation, 1996; Government of Fiji, 2012; Jones, 2013). Yet, activities dependent on externally driven environmental policy tend to exist only for the lifetime of external resourcing (Heckel, 2003; Nunn, 2009), such as waste management projects in Kiribati (Storey and Hunter, 2010). This may be due to poor planning or lack of necessary resources for continuation of the project (Lata and Nunn, 2012). Thus, while donor aid is valuable in providing infrastructure that are otherwise beyond the reach of PICs it must be provided in such a manner to ensure the human and financial resources are available to provide for ongoing operation, maintenance and eventual replacement (Finnigan, 2011).

Employing appropriate technology is therefore a vital consideration for implementation of air quality monitoring in PICs. Equipment that requires a highly stable power supply or strict climate controls may not function effectively in a Pacific Island setting because these are not consistently available. Likewise, if expensive analyses of samples are required as part of the monitoring regime it is unlikely to be continued beyond the timeframe of external funding. Finally, akin to the management of waste compactor vehicles and incinerators (World Health Organisation, 1996), it is necessary that the appropriate financial, technical and personnel resources are available for correct operation and maintenance of air monitoring equipment along with the assessment and interpretation of data.

Further to economic constraints, the infrastructure for waste management in PICs is often inadequate to fulfil the intention of environmental policy. For example, in the Cook Islands, although television advertisements encouraged separation of wastes, waste contractors placed separated refuse and recyclables together in the same truck (Finnigan, 2011). This was because recyclables were being stockpiled rather than exported, reducing incentive for waste separation (Finnigan, 2011). Similarly, the Government of the Federated States of Micronesia (2007, 2010) acted to increase public awareness of the risks of open burning of waste, to enforce burning regulations and provide affordable waste disposal options. Sadly, a lack of infrastructure contributed to poor compliance with these policy measures (Woodruf, 2014b). Whilst waste segregation and recycling initiatives exist in PICs

(Woodruf, 2014a; Anderson, 2016; Mackay, 2017), this lack of appropriate infrastructure means that in the vast majority of cases, waste is collected, dumped on vacant areas, in the ocean or burned (Woodruf, 2014a).

Health risk posed by burning of wastes, or indeed from other environmental risks (such as climate change) is poorly understood in PICs (Government of the Federated States of Micronesia, 2007; Mortreux and Barnett, 2009; Lata and Nunn, 2012). Similarly, motorists are unaware of the potential harm caused by vehicle emissions (Rogo, 2011). A lack of understanding or acceptance of health risk related to air quality is not unique to the Pacific Islands (Environment Protection Authority, 2016). However, the failure to understand the risk associated with inaction on widespread environmental issues, their broader connectivity to human health and the protection of the environment is also shared by decision-makers in PICs (Lata and Nunn, 2012). Ironically, the lack of awareness of societal actions and behaviours has been acknowledged within the Pacific as doing harm to public health (Government of the Federated States of Micronesia, 2007).

3.3. Governance

Communities often resist government directives, deferring to more traditional systems of governance (Keppel et al., 2012a). Amongst the Pacific Islands nations, traditional culture and society are strong, with over 90% indigenous population (Koshy et al., 2008) and over 80% of land under traditional ownership (Keppel et al., 2012b). Further, cities in the Pacific Islands often cross administrative and social boundaries. As urban areas grow, peri-urban settlements develop on customary land; challenging distinctions between the roles of state and customary institutions (Petru, 1993; Pulea, 1994; Storey, 2006; Scaglione, 2013). Land-owning communities believe that making environmental decisions about their land is their traditional right (Dusevic, 2001; Nunn, 2009; Lata and Nunn, 2012); resulting for example, in the refusal to follow directives prohibiting the burning of wastes or crop residues.

Enforcement of environmental policies is complicated by these different legal and governance frameworks. In Micronesia, vast areas of forest were declared as protected areas in 1987, however this was established without consultation of landholders and community leaders and was unsuccessful (Keppel et al., 2012a). A later initiative engaged the existing local leadership, building capacity within the community for self-management of their traditional lands; this has succeeded in reducing erosion and protecting biodiversity (Keppel et al., 2012a; The Nature Conservancy, 2017). Likewise, in Fiji, empowering of existing local governance for management of clam stocks has proven vital to clam conservation (Govan et al., 2012). The implication from these previous challenges for successful implementation of air quality management is that management plans that give traditional leaders power to enforce directives are more likely to succeed (Clarke and Jupiter, 2010). This may involve giving tribal leaders the power to enforce fines for burning rubbish or unauthorised land-clearing by burning, with fines being paid into village funds; noting examples from Tokelau (Angelo, 1993) and Vutia, Fiji (Lata and Nunn, 2012). Land-owning communities should be consulted as to what activities form a vital part of culture and traditional practice. For example, in Tonga, waste management regulations enacted in 2016 (Government of Tonga, 2016) to prevent open burning of wastes still allow burning for some agricultural purposes and traditional cooking in umus (earth ovens).

4. Factors for success

4.1. Increasing awareness

Understanding environmental change and its consequences is key to the successful management of environmental resources (Lauer and Aswani, 2010). Recently, Fiji's 'reduce, reuse, recycle' awareness program (Secretariat of the Pacific Regional Environment Programme,

2016) has seen increased proportions of materials being recycled (and presumably less being burned). In Fiji, the mass of recyclable materials collected increased from 251 t in 2013 to 396 t in 2016 (Waqavonovono, 2017). It is estimated that 57% of Fiji's recyclable waste from vehicles, white goods, cans, PET, paper and cardboard is now exported, reused or recycled (Secretariat of the Pacific Regional Environment Programme, 2016). Surveys to determine the most effective medium for communicating environmental awareness (Lata and Nunn, 2012) indicated radio (31% population reach) to be the key medium, followed by newspapers (25%) and television (22%). Aside from wastes and air quality, turtle conservation provides a further example of a successful public awareness campaign (Fiji Government, 2002; Laveti and MacKay, 2009; Freund, 2014; Secretariat of the Pacific Environment Programme, 2015; CITES, 2017; Fiji Environmental Law Association, 2017b; Solomon, 2017).

4.2. Financial responsibility

For sustained environmental management, PICs need to commit their own resources to local issues (Secretariat of the Pacific Regional Environment Programme, 2016). Before environmentally damaging behaviours can be changed, there needs to be practical, economic and sustainable alternatives. Successful locally-funded initiatives include container deposit programmes (Kiribati, Federated States of Micronesia and Palau), pre-paid waste collection bag systems (Kiribati), and the introduction of waste tipping fees in Fiji (Secretariat of the Pacific Regional Environment Programme, 2016). Other examples include funding environmental management using ecotourism and trust funds (de Haas, 2002; Keppel et al., 2012b).

4.3. A co-ordinated approach

Environmental management and enforcement of policy requires co-ordination between government departments and other groups. It also requires issues to be addressed from multiple angles. Such an approach is well demonstrated by the Fiji's Land Transport Authority's initiative for vehicle emission reduction.

The Fijian Land Transport Authority's plan to combat emissions consist of several steps: Firstly, they are appealing to the public to understand the health dangers of vehicle emissions and to be responsible vehicle owners (Tuinaceva and Shute, 2015). Fines, issued daily, are largely ignored (Land Transport Authority Fiji, 2016); yet recent action to share databases with Fijian courts has enhanced enforcement (Land Transport Authority Fiji, 2016). Further to this, field officers will also receive electronic devices to allow database access (Land Transport Authority Fiji, 2016). Bus emission standards have been set and staff are receiving training about the standards (Prakash, 2016). Also, recently acquired automated motor vehicle inspection systems in Fiji now allow accurate smoke emission testing (Land Transport Authority Fiji, 2016). Change to emissions levels of Fiji's vehicle fleet will likely be gradual; however these actions by the Land Transport Authority seem well placed to foster vehicle emissions reduction in the longer term.

4.4. Regional cooperation

There is a need for regional cooperation to share information and lessons learned, to promote regional and interregional exchange and to undertake joint projects and research activities (Rogo, 2011). Nunn (2009) recommends meetings, both within and between PICs, where communities can detail how they have dealt with environmental issues and learn from each other. The Secretariat of the Regional Environment Programme (2017) facilitates such dialogue between 21 PICs; providing advice and developing regional strategies. This has contributed to significant progress in the way waste is managed; with open dumpsites rehabilitated, garbage collection services expanded and successful

recycling initiatives being implemented in Palau and Kiribati (Secretariat of the Pacific Regional Environment Programme, 2010; Secretariat of the Pacific Regional Environment Programme, 2016).

4.5. Empowering local leadership

Successful environmental management in PICs will include traditionally respected community leaders and stakeholders at every stage of the process: from planning through to ongoing management (Keppel et al., 2012a). Successful projects employing these characteristics include forest conservation in the Solomon Islands (Lauer and Aswani, 2010; Keppel et al., 2012b) and marine and terrestrial resource management in Fiji (Clarke and Jupiter, 2010). Similarly, co-operative waste management programs have developed technical capacity of Pacific Islanders leading toward self-sufficiency in waste management (Secretariat of the Pacific Regional Environment Programme, 2016).

5. Implications for air quality policy

Effective management of wastes and also of vehicles in PICs will bring improvement of air quality. The current extent of air quality issues and their impacts on environment community and health need to be established using relevant local data. Together with an understanding of local community needs and culture, such data will form a starting point for action. As detailed in Section 1, air quality data is largely absent across the PICs. If emissions are to be reduced and air quality improved, a multi-faceted approach involving community awareness-raising, collection of data, provision of alternative options, penalties for pollution and the provision or empowerment of human resources to allow enforcement are required.

5.1. Awareness

In order for air quality policy to be effective air pollution health risks must be effectively communicated to the wider community, especially those in leadership positions. It may be appropriate to invite elders, teachers and religious leaders (Nunn, 2009) to workshops on impacts of air quality and practical ways to reduce emissions at a community level. Ideally, such a workshop would guide the community leaders to develop their own solutions appropriate to their local context (Keppel et al., 2012a); determining how to achieve national air quality goals within their community (Clarke and Jupiter, 2010; Govan et al., 2012). Follow-up sessions may be useful to share successful ideas between communities (Nunn, 2009).

5.2. Data

It is difficult to know that emission reduction has occurred without data. The biggest health risk from particle air pollution identified for Suva City was 'fossil fuel smoke' (Isley et al., 2018a). The study method used in (Isley et al., 2018a) was not able to identify specific industrial premises or other specific point sources for fossil fuel smoke sources. Although estimates of sources were based on emissions inventory data (Isley et al., 2016) from fuel imports, more accurate information needs to be gathered to determine the output from specific industries of interest. Once these are known, it is possible to formulate goals and strategies for improvement; starting with the most significant sources. This emissions inventory method is used by industry and government to reduce air pollution in Australia (Australian Government Department of the Environment and Energy, 2014).

Goals for air quality improvement must be measurable. As most PICs currently lack air quality monitoring, setting air quality standards based on pollutant concentrations are redundant. In this regard it is vital that basic air quality monitoring be established in key locations across the PICs. In residential areas, there is evidence that smoke from biomass and waste burning is a large emission source that represents significant

Table 1

Common elements of successful environmental management strategies in the Pacific Islands. While the fulfilment of each category may vary dependent upon the specific definition of that category, it is noteworthy that these programs/strategies each satisfy a number of these criteria.

Program/ strategy	Section discussed in this study	Locally managed/ empower existing leaders	Locally funded/ co-funded	Public awareness campaign	Co-ordination with other government departments or groups	Capacity building/ training locals
Reduce, reuse recycle	4.1	✓	✓	✓		✓
Turtle conservation	4.1	✓		✓	✓	✓
Container deposit	4.2	✓	✓	✓		✓
Waste tipping fees	4.2	✓	✓			✓
Ecotourism	4.2	✓	✓		✓	✓
Trust funds	4.2	✓	✓		✓	✓
Land transport authority vehicle emissions campaign	4.3	✓	✓	✓	✓	✓
Forest conservation -Solomon Islands	4.5	✓	✓		✓	✓
Marine resource management Fiji	4.5	✓	✓		✓	✓

human health risk (Isley et al., 2016; Isley et al., 2018b). This represents a modifiable risk that needs to be addressed. In addition to air quality monitoring, it may be useful to survey a residential area for burning activity to establish a baseline level and then to repeat the same survey after certain time periods to measure improvement. Vehicle emissions have also been identified as an air pollution source in Fiji that carries a significant health risk (Isley et al., 2018a). Sharing data from the Land Transport Authority, regarding vehicle numbers, emissions test results and frequency of fines issued for excessive smoke, would demonstrate whether vehicle regulations and fuel quality improvements are effective in reducing emissions. For industrial sources, emission reduction should be demonstrated through emissions testing or emissions estimates based on the emissions technology employed.

5.3. Alternatives

Practical and economically feasible alternatives must be available before lasting behaviour change occurs. For waste burning, this means that waste disposal alternatives must be available, even for those living in informal settlements that do not have standard council waste collection. Even where regular waste collection is available in Suva, people still burn their waste (Isley et al., 2016), often because they do not want dogs, wind and rain to disturb their rubbish. Raised waste platforms (imagery Supplementary Section D) are a partial solution to this. A short-term waste levy, used to provide sealed garbage containers for each household may provide another way to remove this barrier. A similar special waste levy was used in Australia to provide containers for green-waste and recyclables collection (NSW Environment Protection Authority, 2017e, 2017g).

Potential to improve the quality of diesel fuels in Fiji, for automotive, industrial and shipping uses should also be investigated, as this would bring a reduction in emissions regardless of other controls (Isley et al., 2016). Diesel emission reduction strategies in PICs may take a similar form to those developed for Australia; which has developed strategies for shipping ports, cruise ships and other diesel emissions (PAE Holmes, 2011; NSW Environment Protection Authority, 2017b, 2017c, 2017h, 2017i).

5.4. Penalties

In regard to control of vehicle emissions, Fiji's Land Transport Authority has commenced a program that holds great potential (detailed in Section 4.3), which should be supported and continued. Improvements to this program may include increased communication with the Department of Environment, in terms of sharing statistics as well as investigation of potential benefits of changes to fuel quality and fuel taxation. As with improvements in air quality, it is critical that results

are measured quantitatively, so that the success of the program can be measured and adjustments made as necessary.

Further to this, penalties for breaching environmental laws need to be realistic and enforceable. As noted above, an important barrier to enforcement of environmental policy is that penalties in PICs for waste burning or vehicle emissions can be too severe. Kiribati faced such an issue (Tiake et al., 2002) with stop or abatement notices being issued to polluters but were not followed through. As a result, it was recommended that on-the-spot fines would be more effective, however these also seem to have been poorly enforced (Sherborne, 2014). Similarly, rather than a \$10,000 dollar fine for burning waste (Government of Fiji, 2007c), an on-the spot fine that represents a few hours earnings may be easier to enforce and more effective in preventing this behaviour. Tonga has recently introduced such regulations (Government of Tonga, 2016). In Australia, a program encouraging the wider community to report the littering activities of others has been highly successful in terms of prosecutions (NSW Environment Protection Authority, 2017a, 2017d, 2017f).

5.5. Human resources

Education programs, waste strategies and gazetted penalties are no use without workers to enforce them. Workers require training and wages, hence environmental management requires financial commitment. The benefits of air quality improvement must be kept in mind; they represent a saving to the community in terms of reduced premature mortality and reduced illness. These carry financial benefits of increased productivity and reductions in health expense (OECD, 2012). A small grants scheme to support community waste-reduction initiatives, such as NSW Environment Protection Authority (2017j), may be appropriate; this would build capacity within communities for waste management without requiring additional government employees.

5.6. Summary

Table 1 demonstrates the common elements between the different successful actions. In particular, these show that they all have a focus on local management and capacity building. Future actions to improve air quality in the Pacific Islands will likely be more effective if they incorporate these previously successful elements.

5.7. Interrelationship with climate change

Interestingly, both the barriers and best practices identified here for successful formation and implementation of air quality policy in the Pacific Islands are similar to those identified by the Pacific Centre for Environment and Sustainable Development (2011) for dealing with

climate change in this region. The interrelationship of particulate air pollutants, especially black carbon, in Fiji with climate change is discussed in (Isley et al., 2016), which quantifies the emissions and hence radiative forcing potential from different emission sources. Hossain (2018) argues that whilst air pollution science typically focuses on health risk and climate science focuses on radiative forcing potential, both issues are based on the burning of biomass and fossil fuels, hence an integrated approach is warranted.

6. Conclusion

The challenges to improving air quality in the Pacific Islands stem from spatially distributed populations, limited resources and infrastructure, failure to understand environmental and health risks as well as complicated systems of governance. Successful environmental management programs in the Pacific have involved the best practices listed in Table 1. These include a high level of local engagement, in terms of planning, financial commitment, development of local leadership and communication between stakeholders; they also incorporate capacity building aspects and frequently make use of public awareness campaigns. For air quality to be accepted as requiring improvement across the Pacific Islands, local authorities must develop an understanding of the current risks. Once this data has been assembled from monitoring and evaluation, it must be communicated effectively to those community leaders who will ultimately be involved in the broader education, dissemination and enforcement of air quality policy.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi: <https://doi.org/10.1016/j.envsci.2018.02.013>.

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Chapter Four: Discussion

Air pollution is the leading global environmental risk factor for disease and premature death (World Health Organization 2014). Evidence for a causal relationship (Brook *et al.* 2013) between exposure and mortality or adverse health effects is strongest for the PM_{2.5} fraction (particles less than 2.5 µm (Choi *et al.* 2004; Franklin *et al.* 2007)). Risk from PM_{2.5} includes cardiovascular (Hoek *et al.* 2013) and all-cause mortality (Kloog *et al.* 2013; Shi *et al.* 2016); with no evidence of a threshold below which effects are not observed (Pope and Dockery 2006; Brook *et al.* 2010). Other associations include diabetes (Potera 2014; Rao *et al.* 2015); deep vein thrombosis and pulmonary embolism (Kloog *et al.* 2015), dementia, Alzheimer's and Parkinson's disease (Kioumourtzoglou *et al.* 2016).

Air quality is a topic of increasing interest in Fiji and other Pacific Islands. The Department of Environment in Fiji aim to reduce air pollution (Department of Environment Fiji 2007; Fiji Department of Environment 2013). Fiji's submissions to the United Nations (Fiji Department of Environment 2010; Rogo 2011; United Nations 2013; Department of Environment Fiji 2015) reflect this. Concern regarding air pollution is common across the Pacific Islands, particularly emissions due to combustion of diesel fuels (Gleye 2010; Dornan and Jotzo 2012; Pacific Energy Summit 2013; Escoffier *et al.* 2016; Taibi *et al.* 2016; Swami 2017) and from waste burning (Thaman *et al.* 2003; Department of Environment Fiji 2007; Periathamby *et al.* 2009; Mataka 2011; Owens *et al.* 2011; Woodruff 2014; Mohee *et al.* 2015). Despite this concern, very little scientific data were available (Chapter One, Section 1.2) to describe air quality or emissions in the region.

This study has observed and defined the particle air quality situation in Suva in great detail. This provides a sound scientific basis for addressing particle air quality and to understand what management options would most effectively reduce preventable exposures. Improving particle air quality brings benefits to the community and the economy in terms of reduced

premature mortality (Krewski *et al.* 2009; Pope *et al.* 2009) and reduced incidence and exacerbation of disease (Laumbach *et al.* 2010). These carry economic benefits of increased productivity and reductions in health expenditure (OECD 2012).

The results of this study not only have implications for Fiji and other Pacific Islands, but also for other small island states affected by open burning of waste (Mohee *et al.* 2015; Marra 2016; Riquelme *et al.* 2016) and reliance on diesel combustion (Mishra *et al.* 2009; Eurelectric 2011; Lucas *et al.* 2017; Wood 2017). Indeed, these combustion sources are significant issues globally for cities in developing countries (Guerrero *et al.* 2013; Mitra 2014; The World Bank 2014; Wiedinmyer *et al.* 2014).

4.1 How aims were addressed:

The principal aim of this study was to investigate particle air quality in the Pacific Islands in order to allow for more effective air quality management. In order to achieve this, this thesis has addressed the following research questions:

1. ***Measurement of the concentration of fine airborne particulates in Suva, ambient and indoor.***

Addressed in Chapter Two and Chapter Three

Results (Paper One) showed that PM_{2.5} concentrations in Suva were close to or likely to exceed relevant criteria. Whilst within the World Health Organization (2006) criteria (10 µg/m³ annual mean), midweek PM_{2.5} in Suva city (mean 8.6 ± 0.4 µg/m³, Paper One) exceed the Australian Government (2016) National Environment Protection (ambient air quality) measure for PM_{2.5} (8 µg/m³ annual mean). Fiji does not yet have an ambient standard for PM_{2.5} (Government of Fiji 2007). Whilst data are limited for Suva's residential areas, Papers One, Two and Six indicate that both Australian and World

Health Organization criteria may be exceeded in areas of Suva. Indoor concentrations of PM_{2.5} (Paper Six) were consistently higher than Suva city results and median values (10.5 µg/m³ over all indoor locations) were in exceedance of these criteria.

Importantly, Papers One, Two and Six also showed significant difference in PM levels across Suva. The largely ocean-influenced area of Suva Point, upwind of the city, experienced consistently lower PM_{2.5} concentrations than the city area. In turn, residential area PM_{2.5} concentrations were consistently higher than the city. This indicated that local sources contributed strongly to PM_{2.5} exposures in Suva.

2. *Exploration of the components of Suva's fine particulates, in terms of carbon content, ultrafine particle components and also in terms of elemental composition.*

Addressed in Chapter Two and Chapter Three

Black carbon was found to comprise approximately one-third of PM_{2.5} mass in Suva city (Paper One). This black carbon concentration (2.2 ± 0.4 µg/m³, mean of Wednesday and Sunday data, Paper One) was high in comparison to global data, being similar to highly industrialised European cities. This indicated that combustion emissions were strongly contributing to Suva's total PM_{2.5}.

Thermally-determined carbon concentrations (Paper Five) supported black carbon results (Paper One), showing high proportions of carbon in Suva's airborne particulate. Papers One, Two and Five all show a similar differential between carbon concentrations in Suva Point (very low concentrations) compared with city and residential areas, again showing the impact of local combustion sources. Paper Five extended the carbon data to include elemental and organic carbon components as well as the percentages of contemporary and fossil derived carbon particulates at each site. Organic carbon (sources discussed in Question 4) was the dominant carbon component. There was a marked difference

between carbon components sampled at Suva city (80% from fossil fuel sources) and the residential area (approx. 50% from contemporary carbon sources), pointing to a difference in particulate emission sources at these locations.

Similarly to black carbon, ultrafine particle concentrations in Suva (Paper Two, mean ambient particle number concentration $1.64 \pm 0.02 \times 10^4 \text{ cm}^{-3}$) were similar to those in much more populated and industrialised cities. Outdoors, ultrafine particle loadings were highest near roadways. Mobile sampling showed that the highest exposures were encountered whilst travelling by bus (mean $10.3 \pm 1.4 \times 10^4 \text{ cm}^{-3}$), which were also elevated by global standards. Together these indicate transport emissions to be a prevalent source of atmospheric particulate for Suva.

Elemental concentrations were determined for a suite of 22 elements for Suva city (Paper Four) and for indoor and outdoor locations across Suva (Paper Six). Again, these indicated differences between locations, indicating differences in the sources of $\text{PM}_{2.5}$ across Suva.

3. Quantification of known emissions to Suva's air and identification of gaps in knowledge.

Addressed in Chapter Three

Emissions estimation was carried out for Suva using government and industry reports as well as survey data and internationally published emissions technique manuals (Paper Three). This indicated diesel fuels to be the largest contributor to both $\text{PM}_{2.5}$ and black carbon emissions. Burning of waste and industrial activities were also substantial

contributors. Paper Three also quantitatively highlighted the contribution of these black carbon emissions in terms of global warming potential.

Due to limited data collection and emissions regulation in Fiji, emissions inventory data were general in nature. That is, estimates for specific industrial premises, traffic and open burning sources could be refined with collection of further data. Still, this inventory (Paper Three) provides an indication of the main sources, showing where further data collection efforts would be of most value. Requiring more detailed reporting from industries and collection of further data concerning traffic volumes, vehicle fleet and open burning practices, would enable a more accurate inventory to be developed.

Paper three quantified potential reductions in $PM_{2.5}$ and black carbon that could be attained in Suva and across the Pacific Islands. These include enforcing controls on waste burning; considering that most households in Suva have council waste collection (Paper Three), this emission source is unnecessary. Also, reducing the sulphur content of diesel fuels in line with global standards and incorporating other emissions controls for industry and vehicles has potential to adduce reductions in $PM_{2.5}$ and particularly in black carbon emissions.

4. Apportionment of Suva's fine particulates to both natural and anthropogenic emission sources.

Addressed in Chapter Three

Paper Four represents a detailed study of the $PM_{2.5}$ sources in Suva's city area. Using elemental concentrations and the statistical method of positive matrix factorisation, nine sources were identified. Natural sea salt particles, formed by wind and wave action

accounted for 25% of city particulate. Of greater interest were the large contributions from fossil fuel burning (21%, including industrial emissions, small shipping vessels and power generation), road vehicles (two distinct sources totalling 17%) and smoke from open burning (7%). Other sources included secondary sulphates, formed in the atmosphere from natural and human-influenced emissions; windblown soil; industrial emissions high in calcium and heavy fuel oil burning (from large ships).

Paper Five supports the identification of fossil fuel combustion products as a major component in city $PM_{2.5}$. The larger contemporary carbon component identified in Suva's residential area adds further insight to these results. It shows that open burning activities and biogenic emissions contribute greater $PM_{2.5}$ mass in Suva's residential areas than was captured in results focussed on the city area (Paper Four). This is important for air quality management, highlighting that air policy must address both fossil fuel combustion emissions as well as those from open burning of wastes.

Paper Five also allows further understanding of Suva's carbon sources. By distinguishing contemporary and fossil components of elemental (EC) and organic carbon (OC), it has shown what proportion of carbon particulates in air are due to fossil fuel combustion (fossil EC and fossil OC); biomass burning (contemporary EC from waste burning, cooking, cigarettes), biogenic aerosols (including pollens and volatile organic compounds from plants, seen as contemporary OC). It also highlighted that secondary organic compounds, both of fossil and contemporary origin, make up at least one-third of carbon mass in Suva's air. This information is useful for air quality management.

Paper Six also indicated substantial variation in sources with location across Suva. In particular, this paper highlighted the role of indoor emissions arising from burning of wood and kerosene for cooking and lighting purposes, which add heavily to indoor PM_{2.5} loadings. Outdoor sources, namely traffic and open burning activities also contributed to indoor PM_{2.5}. Indoor air quality and emission sources were shown to be of importance, due to the large proportion of time spent indoors by Suva residents (Paper Six).

5. *Assessment of the health impacts of particulate air pollutants.*

Addressed in Chapter Three

Health impacts from particulate air pollutants have been well established on a global basis. Paper Four presented a detailed literature review of health risk data for PM_{2.5}, citing evidence that different components, or different emission sources of PM_{2.5}, have disparate toxicity. Further to this, mortality risk was quantified for Suva, defined separately for each PM_{2.5} source. Natural ocean particulate sources, whilst making up 25% of Suva's PM_{2.5} mass, carried a lower health risk. This natural source is unable to be altered. Modifiable combustion sources, including vehicles, industry, shipping and open burning, carried a higher health risk.

Based solely on PM_{2.5} mass, the modifiable sources representing the largest risk were (Paper Four) fossil fuel smoke, road vehicles and waste burning. The higher risk posed by black carbon content caused the health risk to be increased by up to five times compared with risk based on PM_{2.5} mass. Calculations based on black carbon content showed fossil fuel smoke, in Suva alone, to carry a mortality risk of around 59 premature deaths per year. This was significant when compared with the 2012 road toll for all of Fiji, of 41

deaths. Also of importance was the increased risk due to sulphur content, particularly relevant for emissions from open burning.

Paper Six demonstrated that due to the differing $PM_{2.5}$ content and sources across Suva, risk also varied by location. Indoor $PM_{2.5}$ was higher (median 40% higher) than concurrent ambient city concentrations, indicating that risk based on $PM_{2.5}$ mass was greater indoors. Contrary to this, black carbon concentrations indoors were lower (median 44% lower), leading to decreased risk. Households burning wood and kerosene for cooking and lighting had the highest indoor black carbon and sulphur concentrations, markedly increasing health risk.

Paper Five highlighted the increased prevalence of biomass burning (waste burning) in Suva's residential areas. The risk posed by this source is therefore higher in these areas than is accounted for in Paper Four.

6. Provide the relevant government departments and other organisations with the data required to bring about policy change and effectively manage air quality.

Communication has been maintained with the Fijian Ministry of Environment, Ministry of Health, University of the South Pacific and local councils in Suva and Nasinu throughout this work. Drafts of each paper included in this thesis have been sent to the Ministry of Environment for review and final papers have been made available to all stakeholders. Regular meetings were also held with these stakeholders throughout the completion of fieldwork and data analysis, including the presentation of all results in November 2016. Paper Seven provides a framework for management of air pollution sources.

This study has raised awareness of air quality issues in Fiji and the Pacific. As a result, the University of the South Pacific and the University of New Caledonia have apportioned funding to install air quality monitoring equipment in 2018 (Mani 2017). These will continue to monitor PM_{2.5} levels in Suva, Lautoka (Fiji) and Noumea (New Caledonia) as well as to determine elemental concentrations and sources (as per this study). Continued monitoring in Suva allows for measurement of improvement in terms of reduced PM_{2.5} concentrations and reductions in particular emission sources.

4.2 Overall conclusions and implications

The studies provide detailed scientific data on particle air quality in Suva Fiji. Data of this calibre were not previously available for the Pacific Islands, hence this study provides valuable insight into the impacts particularly of diesel burning and open burning of wastes across the region.

Particle air quality is worthy of consideration because it affects people's health. Significant respiratory health issues in Suva (Flynn 1994b; Flynn 1994a; Masoli *et al.* 2004; Patel *et al.* 2008) may be exacerbated by particulate concentrations (Kim 2004; Kim *et al.* 2015), particularly black carbon (Jansen *et al.* 2005; Janssen *et al.* 2011) although Fijian evidence of this linkage is yet to be established. Cardiovascular mortality is the leading cause of death in Fiji (Ministry of Health & Medical Services 2015), followed by diabetes; the risk of these is increased by increased PM_{2.5} concentration (Hoek *et al.* 2013; Potera 2014; Rao *et al.* 2015). Whilst other lifestyle and environmental factors contribute to these conditions (Abraham *et al.* 2014; Kavishe *et al.* 2015) significant reduction in PM_{2.5} is possible for Suva (Paper Three) and provides a way to reduce these exposure pathways and associated risks.

The usefulness of these data is enhanced by the identification and quantification of sources.

The primary modifiable sources of PM_{2.5} and black carbon in Suva (and most likely elsewhere in the Pacific Islands) are diesel burning, road vehicles and waste burning.

Targeting of these emission sources will undoubtedly bring health benefits.

Additionally, these data provide a baseline of particle air quality conditions and source contributions for Suva, against which any progress may be measured. This is useful to gauge the effectiveness of air quality management strategies that are being implemented.

4.3 Implications for the Pacific islands and other Small Island Developing States

Unlike Suva, many areas across the Pacific Islands do not have the same coverage by waste collection services. This includes many cities with high informal settlement population (squatters), notably Honiara, Solomon Islands and Port Moresby, Papua New Guinea (Jones 2013). These cities therefore experience a greater waste burning problem. It follows that PM_{2.5} concentrations are likely to be in exceedance of criteria in these areas and pose a risk to public health (Wiedinmyer *et al.* 2014).

Following publication of Paper Four, an invitation was extended to the World Health Organization's *Thematic Working Group on Air Quality* (Kim 2017). At this regional forum in September 2017, air quality issues facing the Pacific Islands were raised. It was requested that the data generated in this thesis be made available for input to regional air quality and health risk databases (Gumy 2017). These data allow the Pacific Islands to be included in Asia-Pacific regional programs.

In November 2017, participants from the countries of the WHO Western Pacific Region attended a *Member States Consultation Meeting on Addressing the Health Impacts of Air*

Pollution in Manilla included a delegation.. In their report on air quality and health in the country (to be published in June 2018), Fijian delegates listed Papers One, Two and Four of this thesis as some of few quantitative studies done in the Pacific Islands (Morawska 2017). These studies form a valuable baseline for consideration of actions, not only in Fiji, but also within the other Pacific Island countries.

4.4 Limitations

The ambient sampling study conducted here represented only one year of time. Variations in weather and local events can affect air quality (Paper One) and so a larger data set is desired. Further sampling data would also allow more accurate source apportionment (Paper Four). Additionally, any improvements in air quality and reductions in emission sources would be measurable via continued monitoring. As previously mentioned, emissions data available for Fiji and the Pacific Islands are limited. Collection of further source-based emissions data would allow more effective regulation of the most polluting activities.

The Approved Methods for the Sampling Analysis of Air Pollutants in NSW (Department of Environment and Conservation 2007) list guidelines for siting air sampling equipment. These were followed as closely as possible, giving consideration to placing monitoring equipment away from airflow obstructions etc. These Approved Methods allow, however, for consideration of availability and security of sites, both of which were constraints in Suva. The height of the City sampler, which was placed on top of a building, differs from many standard monitoring sites in Australia. This City site was chosen with the aid of the South Pacific Applied Geoscience Commission (SPC), Secretariat of the Pacific Environment Programme and Department of Environment, Fiji. Of the available locations for sampling it was agreed that this site was most suitable as it more clearly represented air quality in the city centre than

other available sites. This site was also considered most suitable as it also served as a meteorological monitoring station for the Australian Bureau of Meteorology, meaning that high-quality wind speed and temperature data were available for comparison to air quality results. Thirdly, this site offered a stable power supply and a high level of security for protection of the sampling equipment. The SPC negotiated the use of this location with the owner, Fiji Ports. The other two fixed monitoring sites were similarly chosen for availability and security. The Suva Point (background) site had previously been used by the University of the South Pacific for air sampling and as such was appropriately set-up for this purpose. The Kinoya site was offered by the landowner to represent the residential area of Suva, after site-usage negotiations failed elsewhere. The landowner at Kinoya kindly allowed installation of electricity in their field (away from the buildings) and assisted with construction of a locked platform for the air sampler; in order to comply with the Approved Methods (Department of Environment and Conservation 2007). The Kinoya and Suva Point sites also offered the benefit of fencing and on-site security personnel. Whilst these sites may not be considered as completely ideal in the Australian regulatory context, they were the best available options for this study.

Similarly, indoor air quality sampling locations were volunteered by residents of the Suva community. It was endeavoured to represent different neighbourhoods across Suva, however, ultimately the choice of locations was limited to those offered. Access to these locations was limited by the resident's availability, meaning that sampling dates and times of day to change filters were restricted. Combined with availability of equipment, this meant that studies of indoor air quality were limited to a few weeks or less in each location. Exact location of samplers within the resident's home (detailed in Table 1 of Paper Seven) was also subject to approval by the resident. Differences in housing design and location of some samplers in semi-enclosed spaces (which are not either strictly indoor or outdoor) would also affect

comparability of results. This means that results between locations are not directly comparable to each other, seasonal meteorological differences or short-term pollution events may also affect results. It was also not possible to co-locate an outdoor sampler (for indoor-outdoor comparison) at each of these indoor sites, due to security constraints.

Overall, these data indicated that indoor $PM_{2.5}$ may exceed health-based air quality standards (ambient), particularly where there is indoor usage of kerosene and biomass fuels.

Differences in household cooking practices, housing construction and other household activities, which cannot be fully accounted for in this study, would cause differences in indoor air quality and exposures (Buonanno *et al.* 2014; Pacitto *et al.* 2018). A more detailed sampling study is required to fully delineate indoor air quality risks in the Pacific Islands. This is important considering that 40% of Fijian households use biomass fuels for cooking (United Nations Environmental Programme 2015).

Ultrafine particle measurements were constrained by sampler availability (only one sampler, available for a limited time period), meaning that only one location at a time was able to be sampled. This again creates difficulties in directly comparing results from different sites, providing more of an indicative analysis to highlight locations and pollutant sources that warrant further investigation. It was clear from these limited analyses that transport emissions are a significant consideration for air quality and pollutant exposures in Suva. Children's exposures differ from adults (Clifford *et al.* 2016; Goldizen *et al.* 2016; Kurt *et al.* 2016), both due to differences in how they spend their time and due to their physiology. The ethics approval for this project did not include working with children; hence childhood exposure was not investigated.

Differences between air quality monitors that measure different particle sizes and each have different constraints and accuracies create some difficulty in direct comparison if results

between the different published studies included in this thesis. These different measurement devices were co-located wherever possible, in order to provide a measure of their comparability; with equipment comparison data included throughout the published studies in this thesis.

Results for surveys conducted as part of this thesis were also limited by the willingness of Suva residents to respond. The margin of error reported for survey results throughout the papers in this thesis was calculated according to Cornell University (2018). Demographic questions were included at the beginning of the survey and were used to ensure an appropriate sample of age ranges, household sizes and residential locations were included.

Air quality calculations throughout this thesis rely on accurate measurement of flowrate and gravimetric mass of samples. Specifications for the portable air samplers SKC (SKC 2005; SKC 2018) and Microvol (Ecotech 2018), complying with Australian standard for PM_{2.5} measurement AS/NZS 3580.9.10 2006, are included as Appendix G. Samplers were calibrated before and after use in the laboratory using a Gillan Gillibrator-2 primary flow calibrator (Appendix G), as per the Mine Safety and Health Administration (2014). In the field, actual flow volume was measured using a Shigawa DC-1 dry test gas meter (Appendix G). Correlation of SKC pumps with other samplers is detailed in (Zhou *et al.* 2017).

Use of heated inlets on the fixed Osiris samplers counteracted humidity effects on measurements; however, this also results in a loss of volatile components (Deary *et al.* 2016). Hence Osiris data may underestimate the total particulate load in Suva's air.

After at least 24 hours equilibration to laboratory conditions, samples were processed in the ion beam analysis laboratory at the Australian Nuclear Science and Technology Organisation (ANSTO), using a microbalance capable of gravimetrically weighing to ± 1 pg. Before and after each weighing session, this was calibrated with standard weights of 20, 50, 100 and 200

mg. Measurements in this laboratory comply with the Approved Methods for the Sampling Analysis of Air Pollutants in NSW (Department of Environment and Conservation 2007). Accuracy for gravimetric mass in this laboratory was ± 3 pg for a total mass of between 50 and 500 pg of material per filter, including temperature and humidity variation impacts, as detailed in (Cohen 1996).

Whilst Suva is, in many aspects, representative of areas across the Pacific Islands, local differences would also impact on air quality. Extending monitoring to other Pacific Islands is therefore desirable.

Health risk assessment (Paper Four) considered only mortality risk. In reality, health risk and the impact of air quality on society is much broader than this. No consideration was given to hospital presentations, days missed from school or employment, doctor's visits, medication requirements and other less severe symptoms. Improvements to air quality have a wider impact on the quality of life in the community as a whole.

4.5 Future directions

The main recommendations of this study are firstly that the modifiable emissions sources that carry the highest health risk be addressed. These include fossil fuel burning (industrial and shipping), road vehicle emissions and open burning. Secondly, monitoring should continue to allow ongoing assessment of air quality and as an avenue to measure progress.

Diesel combustion emissions be addressed

Electricity generation, industry and shipping in Suva use poor-quality, high-emission fuels. Improvement in fuel quality would bring measurable emission reductions (Paper Three). The same is true for automotive diesel fuels, which currently contain up to 500 ppm sulphur. Fuel

quality is however only one of the means to address these emissions. Emissions controls on industrial processes and on vehicles also need to be pursued. In regard to road vehicles, Fiji's Land Transport Authority Fiji (2016) has begun a program aimed at reducing vehicle emissions. This co-ordinates databases with field officers and the court system and includes an awareness campaign, training, emissions standards and vehicle emissions testing (Paper Seven). This represents a positive step toward effective air quality management in the Pacific Islands and needs to be supported. Collection of data regarding vehicle emissions and fines will enable the monitoring of progress in regard to emissions from Fiji's vehicle fleet. For non-road vehicles and marine vessels, methods outlined in the New South Wales Environment Protection Authority (2017) Diesel and Marine Emissions Management Strategy should be considered. Whilst developed for Australia, these strategies may prove useful in the Pacific Islands context. Of particular interest are goals for reducing shipping emission impacts on urban areas close to ports, which may involve use of lower-sulphur fuels for near-port activities.

Open burning emissions be addressed

In Suva, waste burning is largely an unnecessary activity (Paper Three), yet it contributes significantly to emissions (Paper Four and Five). Addressing open waste burning has the potential to bring benefits to air quality and health in Fiji. Fiji's open fires by-laws (Government of Fiji 2007) include a \$10,000 (FJD) penalty for burning household garbage without a permit. This penalty is severe, being half of the average annual wage in Fiji and is not enforced (United Nations Environmental Programme 2015). Instead, surveys show that waste burning is widespread in Fiji; over 50% of people in Suva (capital) burn household waste and greenwaste ((McDowall 2005; Department of Environment Fiji 2007; Isley *et al.* 2016). Likewise, for Micronesia (Government of the Federated States of Micronesia 2007), Kiribati (Kiribati 1999; Aitken 2014) and the Marshall Islands (Marshall Islands 1984;

Secretariat of the Pacific Environment Programme 2013); enforcement of bylaws and policy around waste disposal is minimal and are ineffective at preventing burning and illegal dumping of waste. Setting realistic penalties and enforcing them would assist to reduce open burning emissions. For example, Tonga's recently introduced open burning regulations (Government of Tonga 2016) set realistic fines for waste burning whilst still allowing traditional cooking activities. Considering that many people burn waste to prevent wind and dogs dispersing it (Pannapadipo 2005; Ehringer 2017), a campaign to introduce affordable and sturdy rubbish bins may be of benefit. As previously mentioned, not all Pacific Island cities have the waste collection facilities enjoyed by Suva residents. Development of waste management programs will be necessary to mitigate waste burning in these areas (Paper Seven).

Air quality monitoring

There are plans to continue particulate air quality monitoring in Fiji (Mani 2017). This locally funded monitoring is a positive response to the results of the papers in this thesis. This monitoring should continue to investigate the concentrations of PM_{2.5} and the sources that contribute to them, in order that air quality and health may be continuously improved.

It is important that particulate air quality monitoring also be carried out to characterise other Pacific Island environments. This should include assessment of indoor air quality, in order to identify the major emission sources and minimise health risks.

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Zhou, X., Aurell, J., Mitchell, W., Tabor, D., Gullett, B., 2017. A small, lightweight

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<https://doi.org/10.1016/j.atmosenv.2017.01.029>

Appendices

Appendix A: Ethics approvals Australia and Fiji

Appendix B: Supplementary Information for Paper Two

Appendix C: Supplementary Information for Paper Four

Appendix D: Supplementary Information for Paper Five

Appendix E: Supplementary Information for Paper Six

Appendix F Supplementary Information for Paper Seven

Appendix G Portable samplers and flow calibration meters

Appendix A:

Ethics approvals Australia and Fiji

Dear Professor Nelson,

RE: Ethics project entitled: "Investigation of Air Quality in Suva, Fiji"

Ref number: 5201400557

The Faculty of Science Human Research Ethics Sub-Committee has reviewed your application and granted final approval, effective 23rd June 2014. You may now commence your research.

This research meets the requirements of the National Statement on Ethical Conduct in Human Research (2007). The National Statement is available at the following web site:

<http://www.nhmrc.gov.au/files/nhmrc/publications/attachments/e72.pdf>.

The following personnel are authorised to conduct this research:

Professor Peter Nelson
Mrs Cynthia Faye Isley

NB. STUDENTS: IT IS YOUR RESPONSIBILITY TO KEEP A COPY OF THIS APPROVAL EMAIL TO SUBMIT WITH YOUR THESIS.

Please note the following standard requirements of approval:

1. The approval of this project is conditional upon your continuing compliance with the National Statement on Ethical Conduct in Human Research (2007).
2. Approval will be for a period of five (5) years subject to the provision of annual reports.

Progress Report 1 Due: 23rd June 2015

Progress Report 2 Due: 23rd June 2016

Progress Report 3 Due: 23rd June 2017

Progress Report 4 Due: 23rd June 2018

Final Report Due: 23rd June 2019

NB. If you complete the work earlier than you had planned you must submit a Final Report as soon as the work is completed. If the project has been discontinued or not commenced for any reason, you are also required to submit a Final Report for the project.

Progress reports and Final Reports are available at the following website:

http://www.research.mq.edu.au/for/researchers/how_to_obtain_ethics_approval/human_research_ethics/forms

3. If the project has run for more than five (5) years you cannot renew approval for the project. You will need to complete and submit a Final Report and submit a new application for the project. (The five year limit on renewal of approvals allows the Committee to fully re-review research in an environment where legislation, guidelines and requirements are continually changing, for example, new child protection and privacy laws).

4. All amendments to the project must be reviewed and approved by the Committee before implementation. Please complete and submit a Request for Amendment Form available at the following website:

http://www.research.mq.edu.au/for/researchers/how_to_obtain_ethics_approval/human_research_ethics/forms

5. Please notify the Committee immediately in the event of any adverse effects on participants or of any unforeseen events that affect the continued ethical acceptability of the project.

6. At all times you are responsible for the ethical conduct of your research in accordance with the guidelines established by the University. This information is available at the following websites:

<http://www.mq.edu.au/policy/>

http://www.research.mq.edu.au/for/researchers/how_to_obtain_ethics_approval/human_research_ethics/policy

If you will be applying for or have applied for internal or external funding for the above project it is your responsibility to provide the Macquarie University's Research Grants Management Assistant with a copy of this email as soon as possible. Internal and External funding agencies will not be informed that you have final approval for your project and funds will not be released until the Research Grants Management Assistant has received a copy of this email.

If you need to provide a hard copy letter of Final Approval to an external organisation as evidence that you have Final Approval, please do not hesitate to contact the Ethics Secretariat at the address below.

Please retain a copy of this email as this is your official notification of final ethics approval.

Yours sincerely,
Richie Howitt, Chair
Faculty of Science Human Research Ethics Sub-Committee
Macquarie University
NSW 2109



**MINISTRY OF EDUCATION, NATIONAL HERITAGE,
CULTURE & ARTS**

Quality Education for Change, Peace and Progress



Resident Address: Marela House, 19 Thurston Street, Suva, Fiji.

Postal Address: Private Mail Bag, Government Buildings, Suva, Fiji.

Ph: (679) 3314477

Fax: (679) 3303511

Our Reference: RA20/14

Date: 28th April 2014

Ms Cynthia Isley
Macquarie University
Australia.

Re: **Official Approval to Conduct Research in Fiji**

Dear Ms Isley

We are pleased to inform you that the approval for the request to conduct research in Fiji has been granted on the topic "Investigation of air pollutant emission and air quality in Suva Fiji; through use of inventory, monitoring and modeling."

The approval is granted from April 2014 to October 2017 as specified in your request.

It is also noted that in this research, you will be working closely with the Department of Environment Fiji, who would be assisting you with facilitating your research. Please liaise with the relevant personnel and organizations with regards to the logistics and the conduct of your research and be further advised that the Government of Fiji's legislations, procedures, policies and protocols must be unreservedly adhered to.

As a condition for all research approvals, a copy of the final research paper must be submitted to this office upon completion. This will be reserved in the MoE Research Library and will be availed for reference by Senior Ministry and Government officials.

Moreover, it is important to note that the Ministry of Education reserves a right to publish the final report or an edited summary of it.

Please liaise with the Immigration Department in regards to the issuance of your Research Permit.

We further wish you success in your research project.

.....
Parmeshwar Mohan (Mr)

for **Permanent Secretary for Education, National Heritage, Culture & Arts**

cc. MoE Research File
Department of Environment

ALL COMMUNICATIONS TO BE ADDRESSED TO THE PERMANENT SECRETARY FOR EDUCATION,
NATIONAL HERITAGE, CULTURE & ARTS

Appendix B::

Supplementary Information for Paper Two

**Airborne ultrafine particles in a Pacific Island Country: characteristics,
sources and implication on human exposure**

C.F. Isley, P.F. Nelson, M.P. Taylor, M. Mazaheri, L. Morawska A. J. Atanacio,

E. Stelcer, D. D. Cohen, Anthony L. Morrison

Section A: Figures and Tables

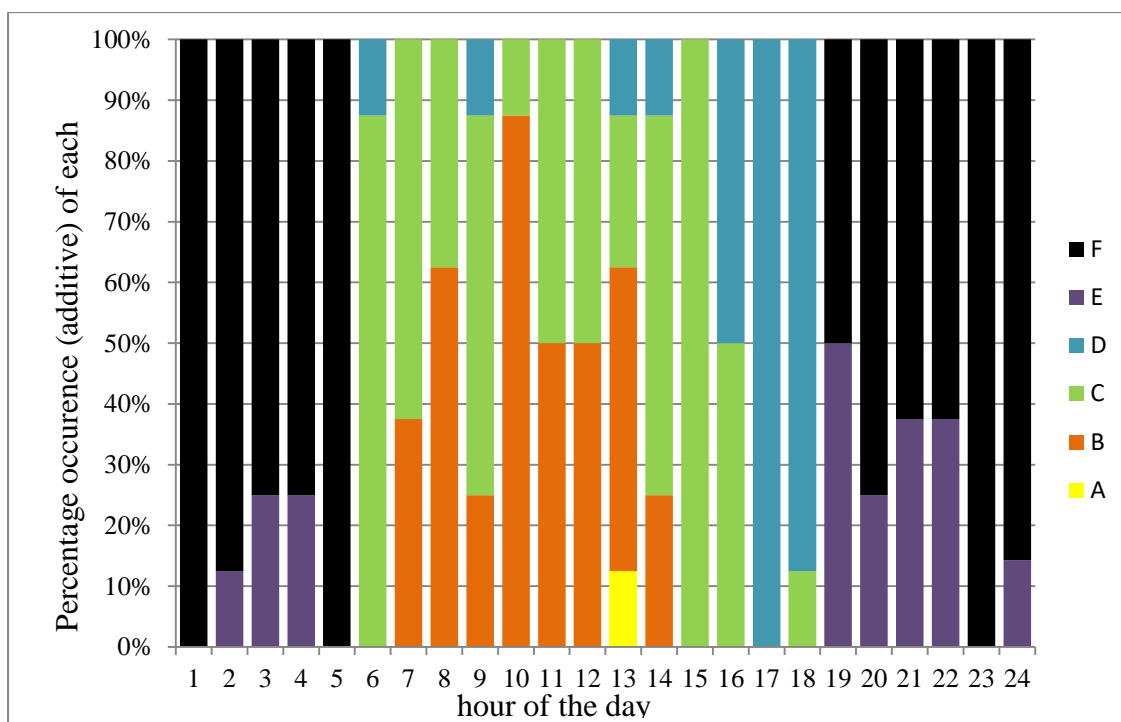


Figure S1: Pasquill Guildford stability classes for Suva, averaged over the 8-day monitoring period to give a typical daily occurrence profile; Class A is the most unstable, class F the most stable.

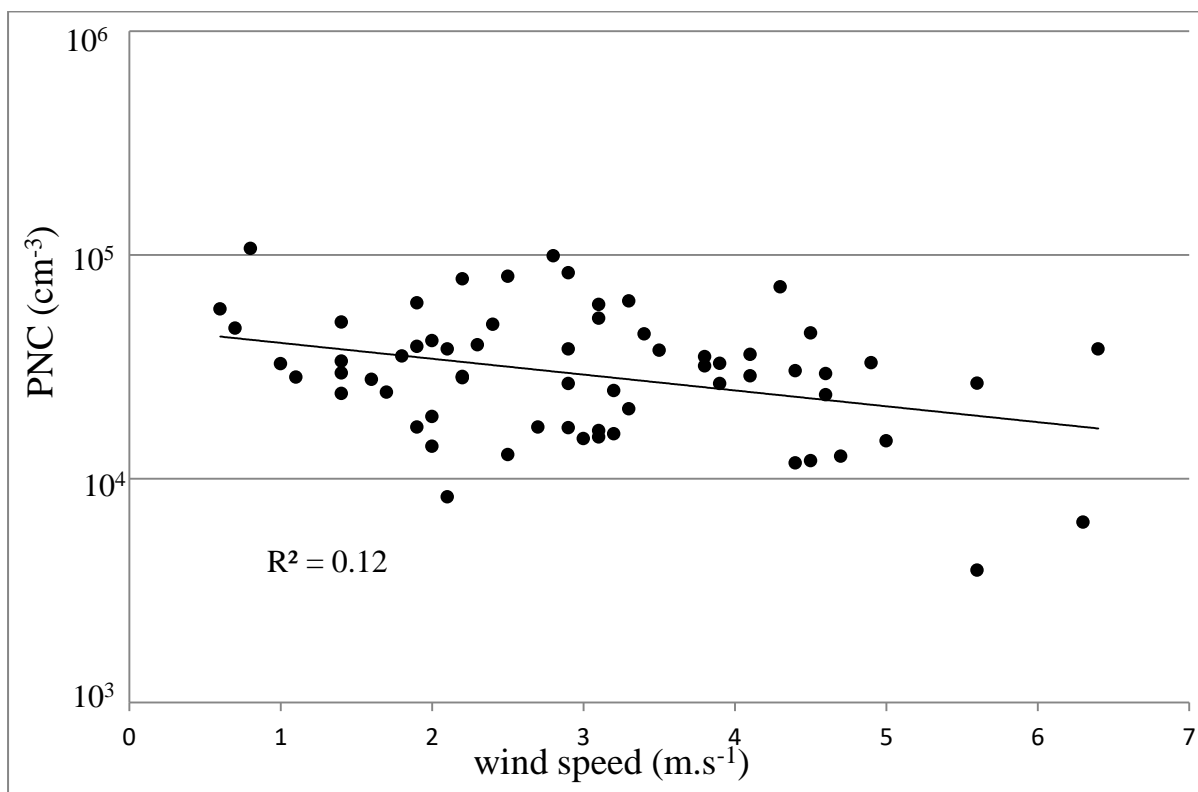


Figure S2: Wind speed v. PNC at Suva City, 1-minute averaged data

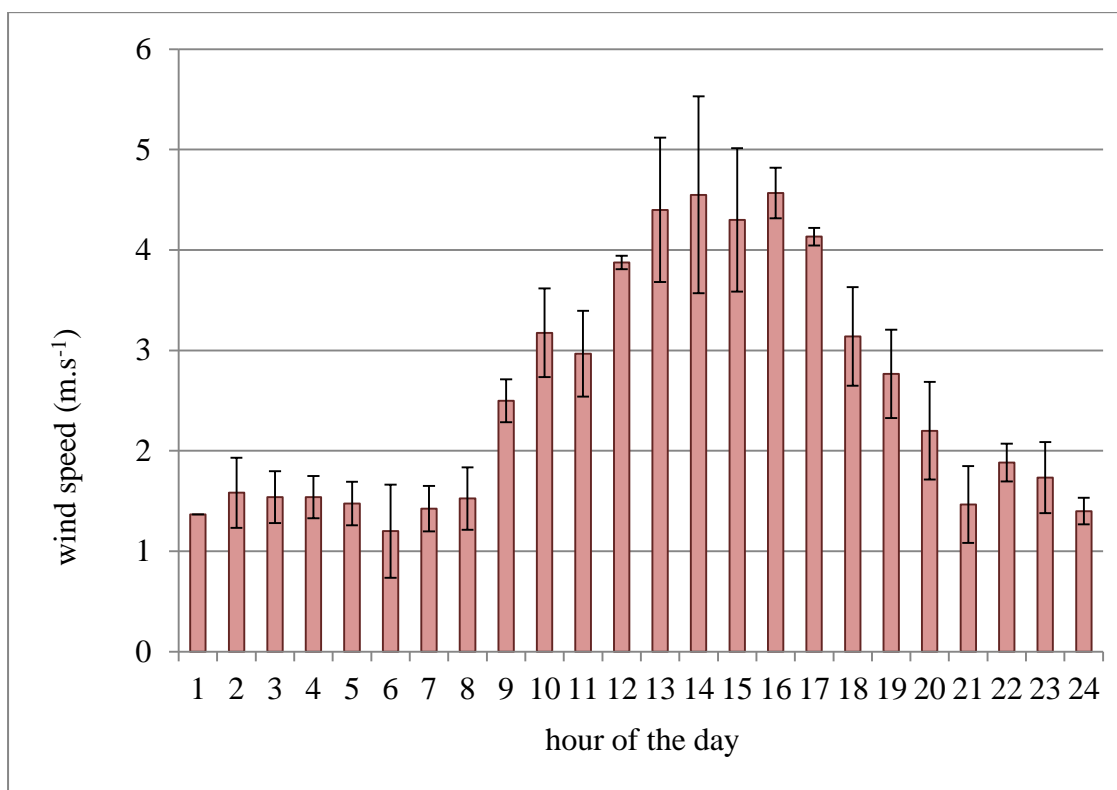


Figure S3: Mean wind speed by hour of the day during sampling at Suva City, averaged over 8 days

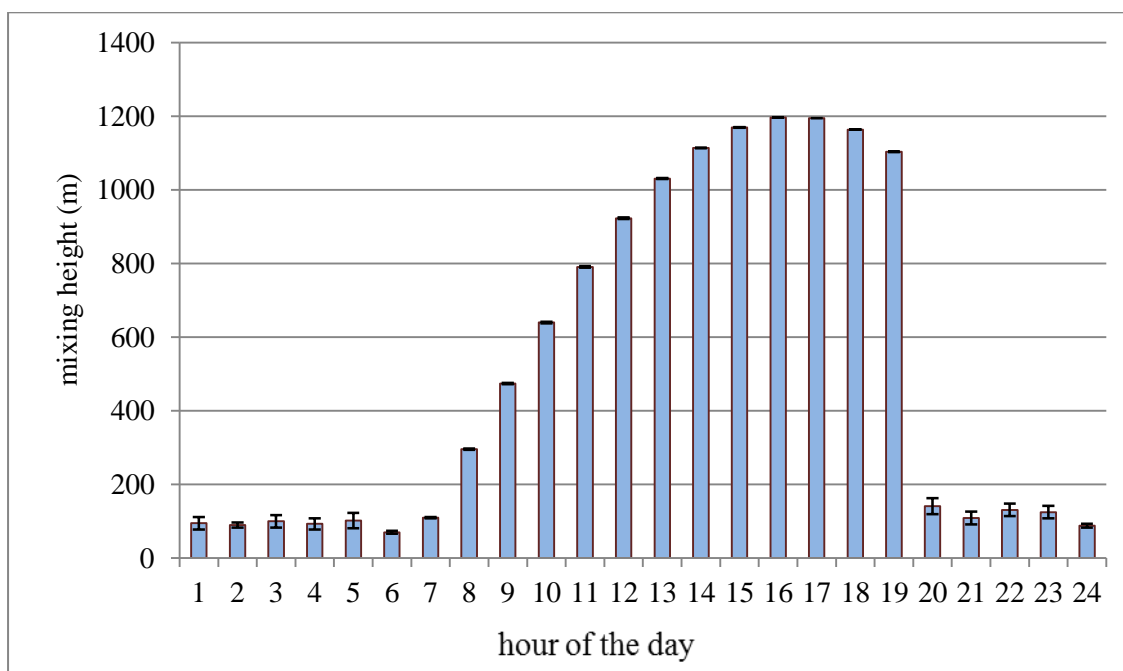


Figure S4: Atmospheric mixing height by hour of day at Suva City, averaged over 8 days

Table S1: PNC and PM data for Suva Point and Kinoya, showing data from the October 2015 study period and other available data for these sites.

	Suva Point		Kinoya	
	October 2015 period	Previous data ¹	October 2015 period	Previous data ¹
PNC x 10 ⁴ cm ⁻³	0.148 ± 0.004	-	0.44 ± 0.02	-
Dp nm	26.2 ± 0.3	-	33.2 ± 0.4	-
HVAS TSP µg/m ³	-	25 ± 3	-	49 ± 11
Osiris TSP µg/m ³	36.2 ± 2.4	9.1 ± 0.1	-	44.2 ± 0.4
Osiris PM ₁₀ µg/m ³	11.4 ± 0.7	4.80 ± 0.04	-	21.7 ± 0.3
Osiris PM _{2.5} µg/m ³	11.4 ± 0.7	3.80 ± 0.02	-	11.8 ± 0.1
Osiris PM ₁ µg/m ³	2.1 ± 0.3	1.30 ± 0.01	-	3.60 ± 0.04
Microvol TSP* µg/m ³	-	26 ± 4	-	39 ± 3
Microvol PM _{2.5} µg/m ³	-	5.5 ± 0.8	-	12.7 ± 1.5
Microvol BC µg/m ³	-	0.2 ± 0.2	-	1.4 ± 0.2

1. Previous data are from October 2014- October 2015 except high volume air sampling, which occurred over a four-year period (Garimella and Deo 2007). Kinoya Microvol 8 TSP and 6 PM_{2.5} samples October 2014 and April 2015, Suva Point 13 TSP and 7 PM_{2.5} samples October 2014 and April 2015. Previous Osiris data are from (Isley *et al.* 2017).

References:

- Garimella, S., Deo, R. N., 2007. Neutron activation analysis of atmospheric aerosols from a small Pacific Island country: a case of Suva, Fiji Islands. *Aerosol and Air Quality Research* 7, 500-517.
- Isley, C. F., Nelson, P. F., Taylor, M. P., Mani, F. S., Maata, M., Atanacio, A., . . . Cohen, D. D., 2017. PM_{2.5} and aerosol black carbon in Suva, Fiji. *Atmospheric Environment* 150, 55-66.
10.1016/j.atmosenv.2016.11.041

Appendix C:

Supplementary Information for Paper Four

Reducing mortality risk by targeting specific air pollution sources:

Suva, Fiji

C. F. Isley, P. F. Nelson, M.P. Taylor, E. Stelcer, A. J. Atanacio,

D. D. Cohen, F.S. Mani and M. Maata

Section A: Supplementary Figures

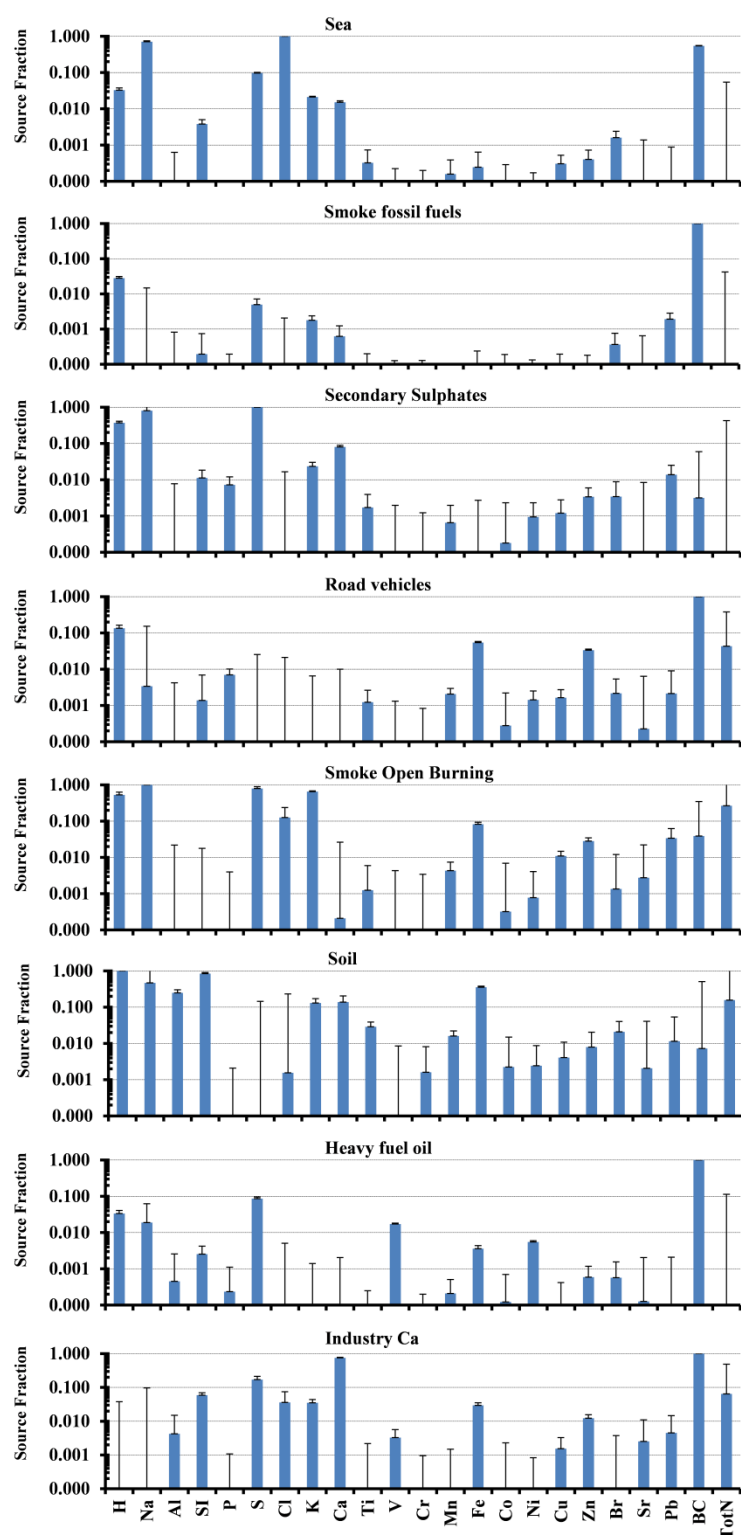


Figure A1: Eight-factor PMF model results. Factors identified at Suva city, showing contribution of each factor to total PM_{2.5} mass. The vertical axis of each graph has been normalised so that the main element in each factor has a fractional contribution value of one.

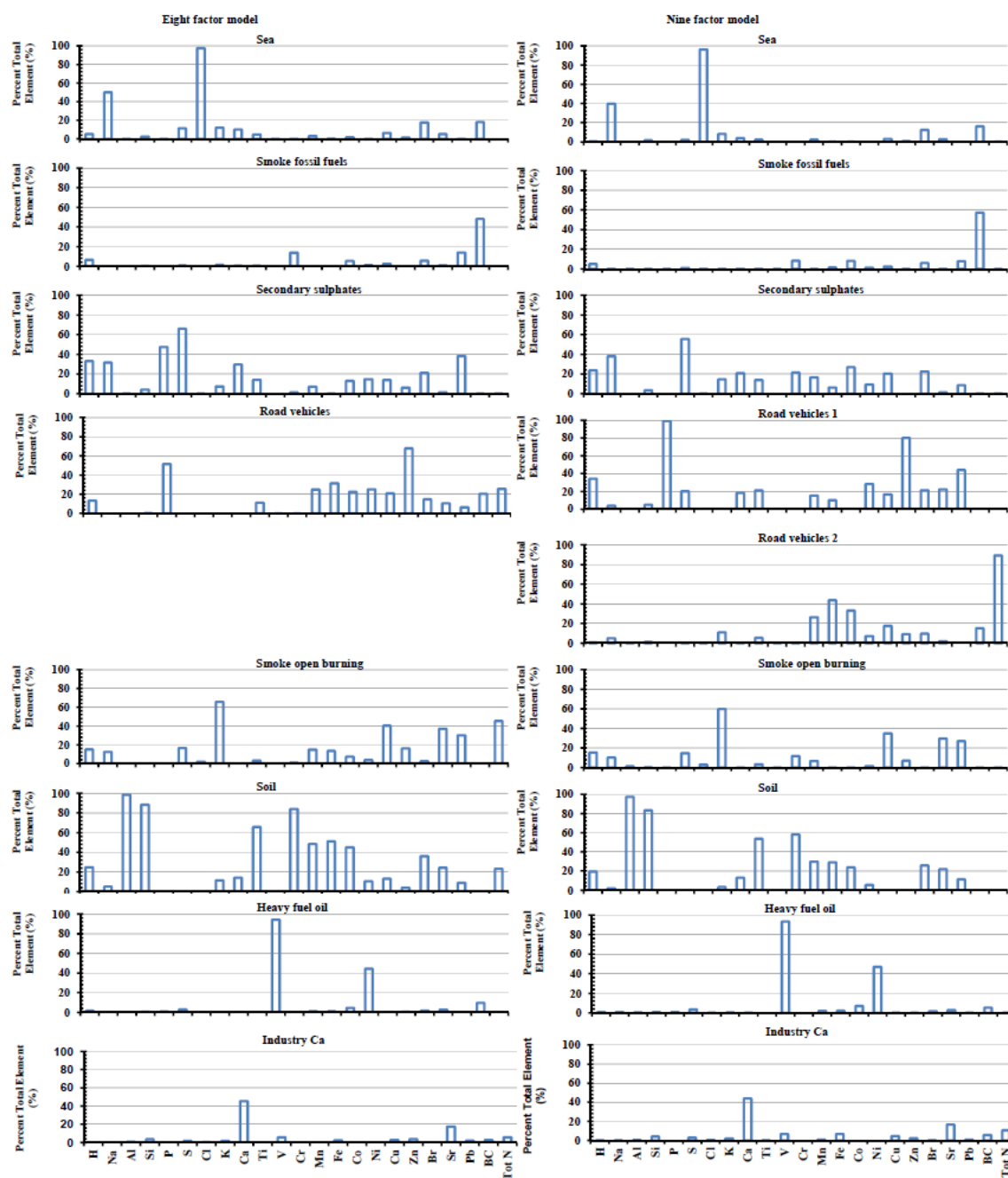


Figure A2: Percentage of total mass of each element, as determined by ion beam analysis, apportioned by factor, for both eight and nine factor PMF models. Results are for PM_{2.5} samples collected in Suva city during 2014 and 2015.

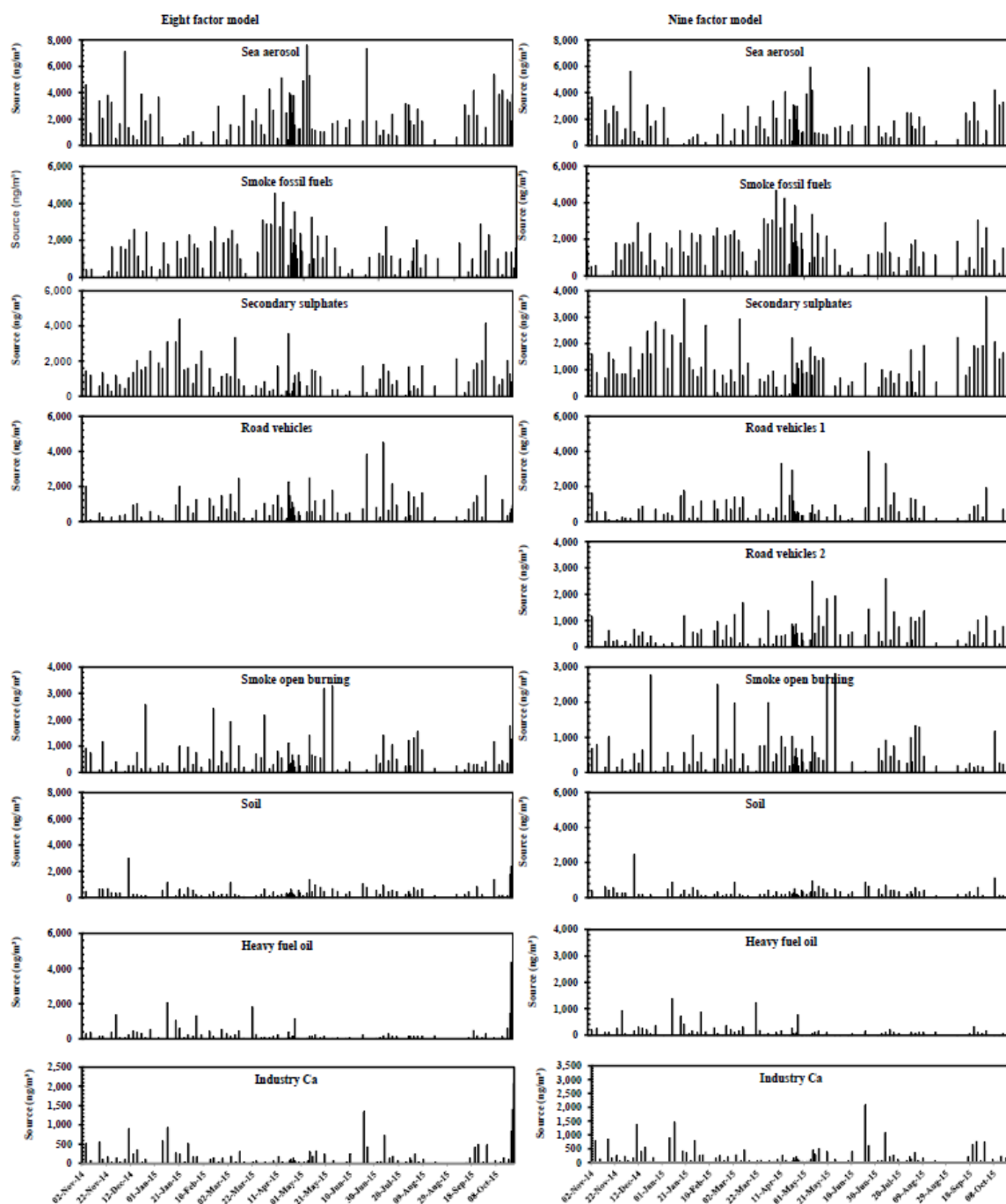


Figure A3: Daily occurrence of each source factor as determined by eight and nine factor PMF models for $PM_{2.5}$ samples collected in Suva city during 2014 and 2015. Elemental concentrations were determined by ion beam analysis.

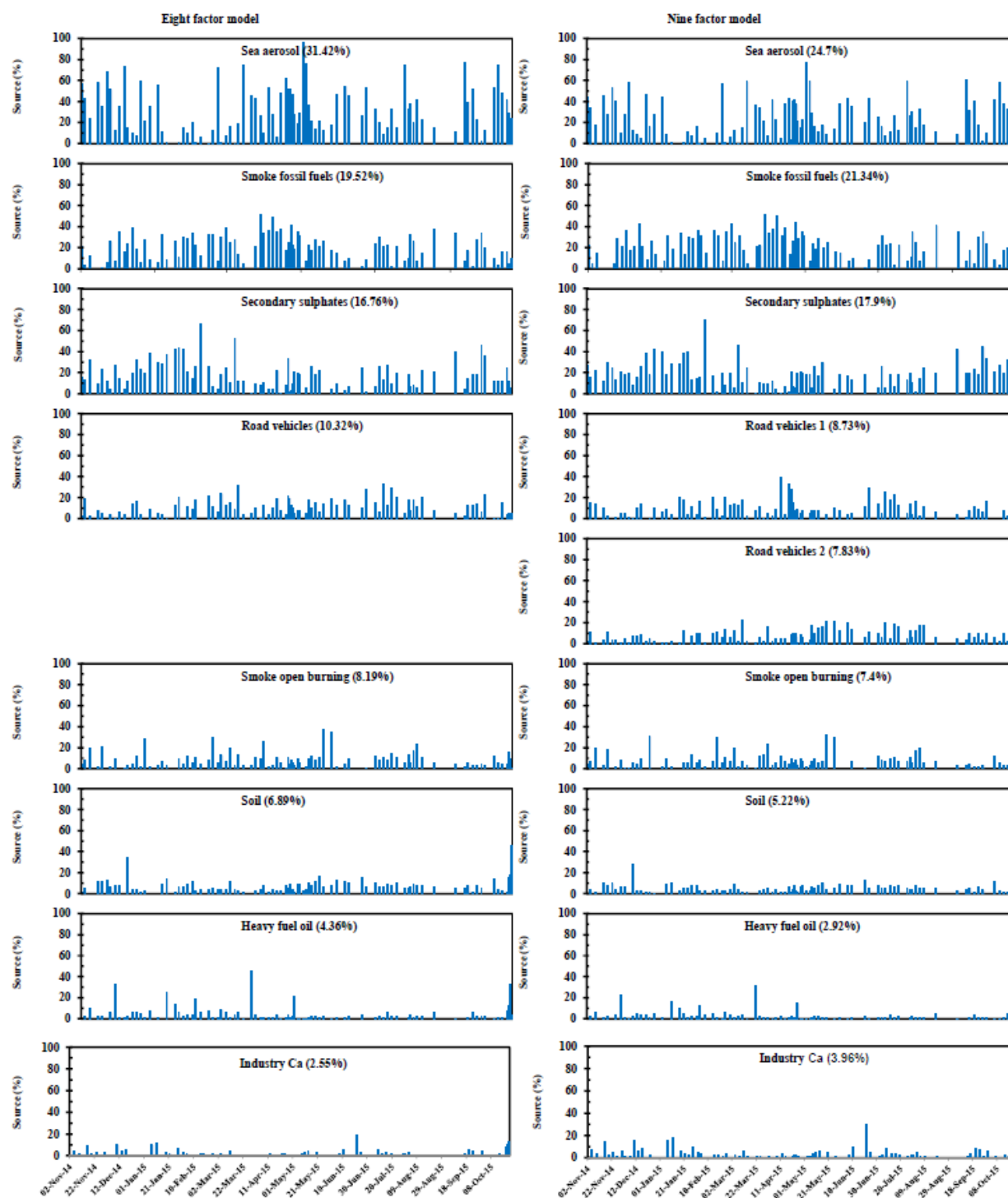


Figure A4: Daily occurrence of each source factor as a percentage of total $PM_{2.5}$ mass; determined by eight and nine factor PMF models for $PM_{2.5}$ samples collected in Suva city during 2014 and 2015. Elemental concentrations were determined by ion beam analysis.

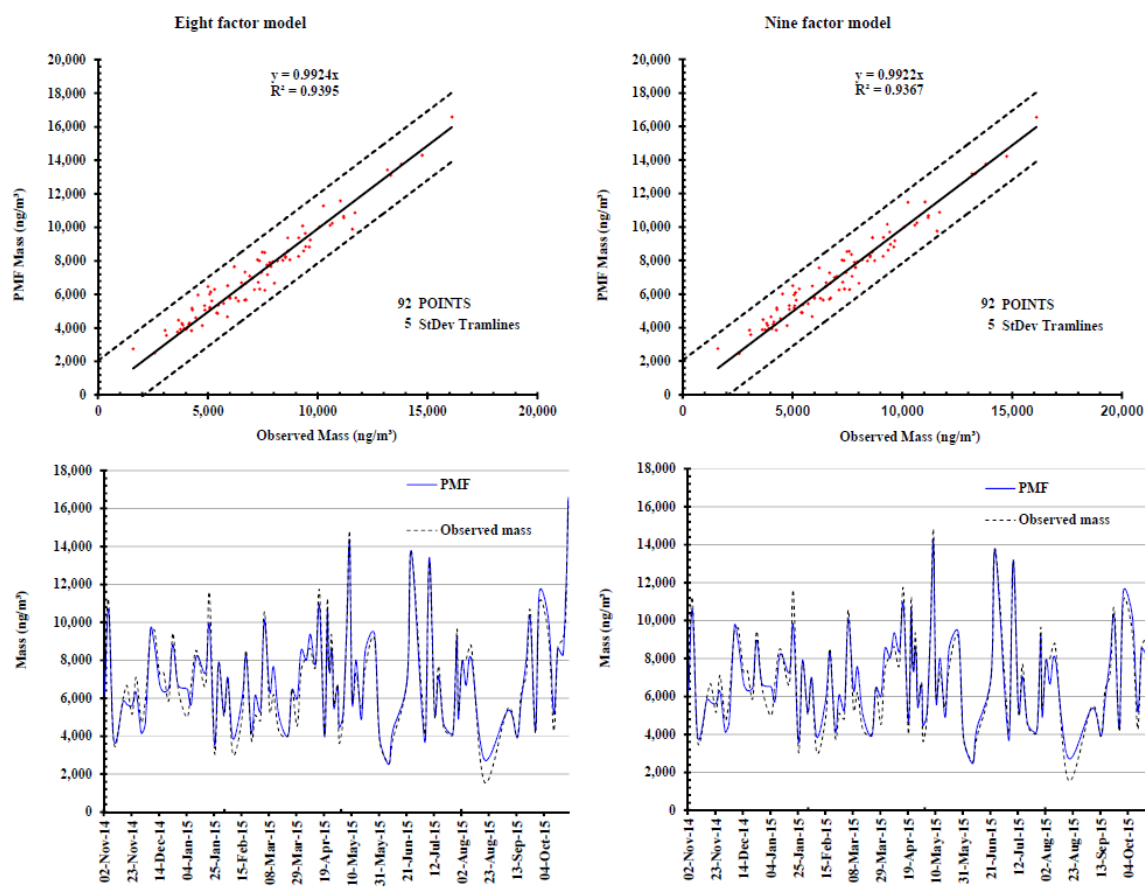


Figure A5: Mass plots for both the eight and nine factor PMF models for Suva city PM_{2.5}. Graphs show the agreement between observed mass and mass determined by PMF for all samples (top) and as a time series (bottom). Results are for PM_{2.5} samples collected in Suva city during 2014 and 2015.

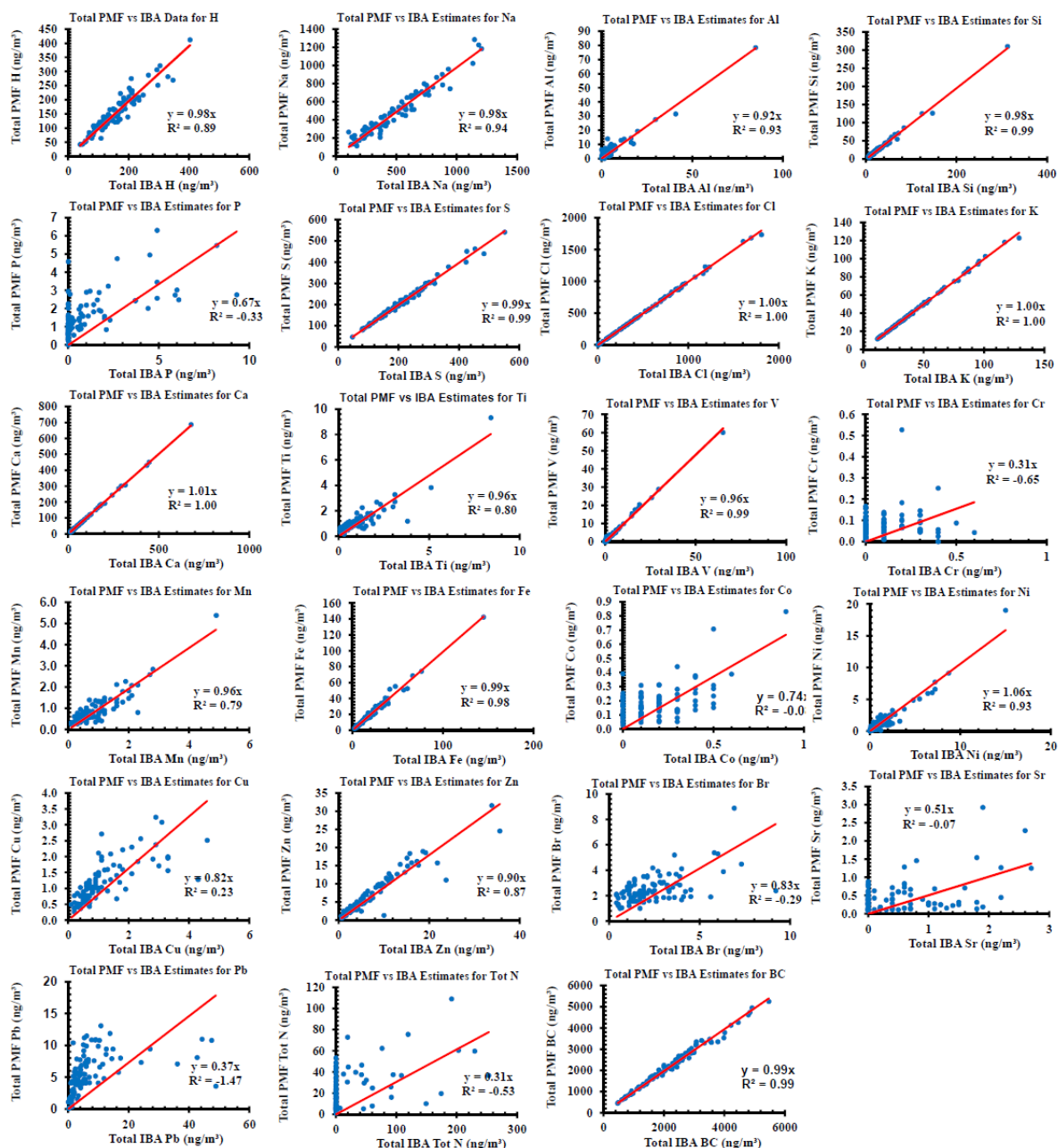


Figure A6: Mass plots for the eight factor PMF model for Suva city $PM_{2.5}$. Graphs show the agreement between mass determined by ion beam analysis and mass determined by PMF for each element. Results are for $PM_{2.5}$ samples collected in Suva city during 2014 and 2015.

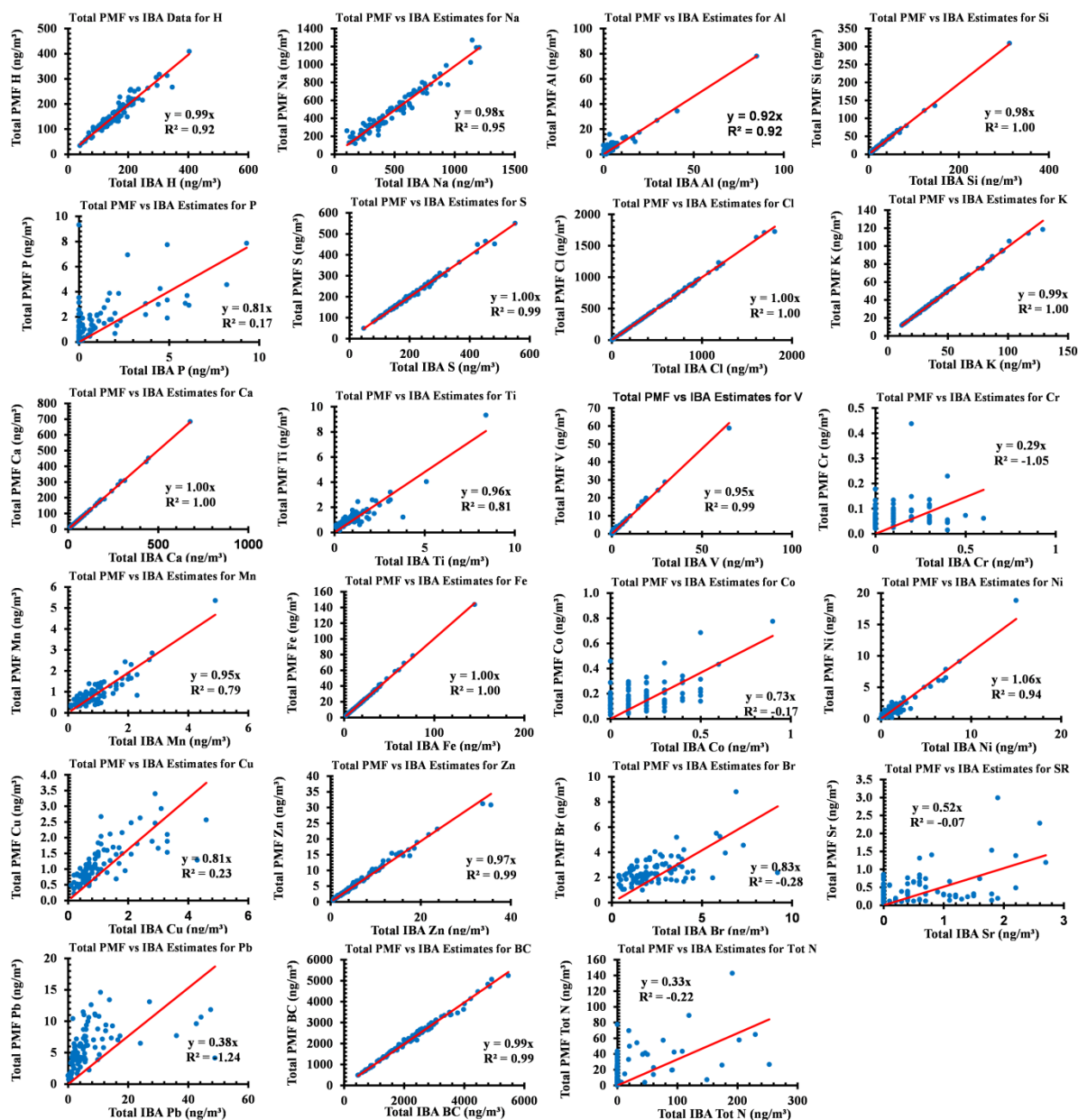


Figure A7: Mass plots for the nine factor PMF model for Suva city $PM_{2.5}$. Graphs show the agreement between mass determined by ion beam analysis and mass determined by PMF for each element. Results are for $PM_{2.5}$ samples collected in Suva city during 2014 and 2015.

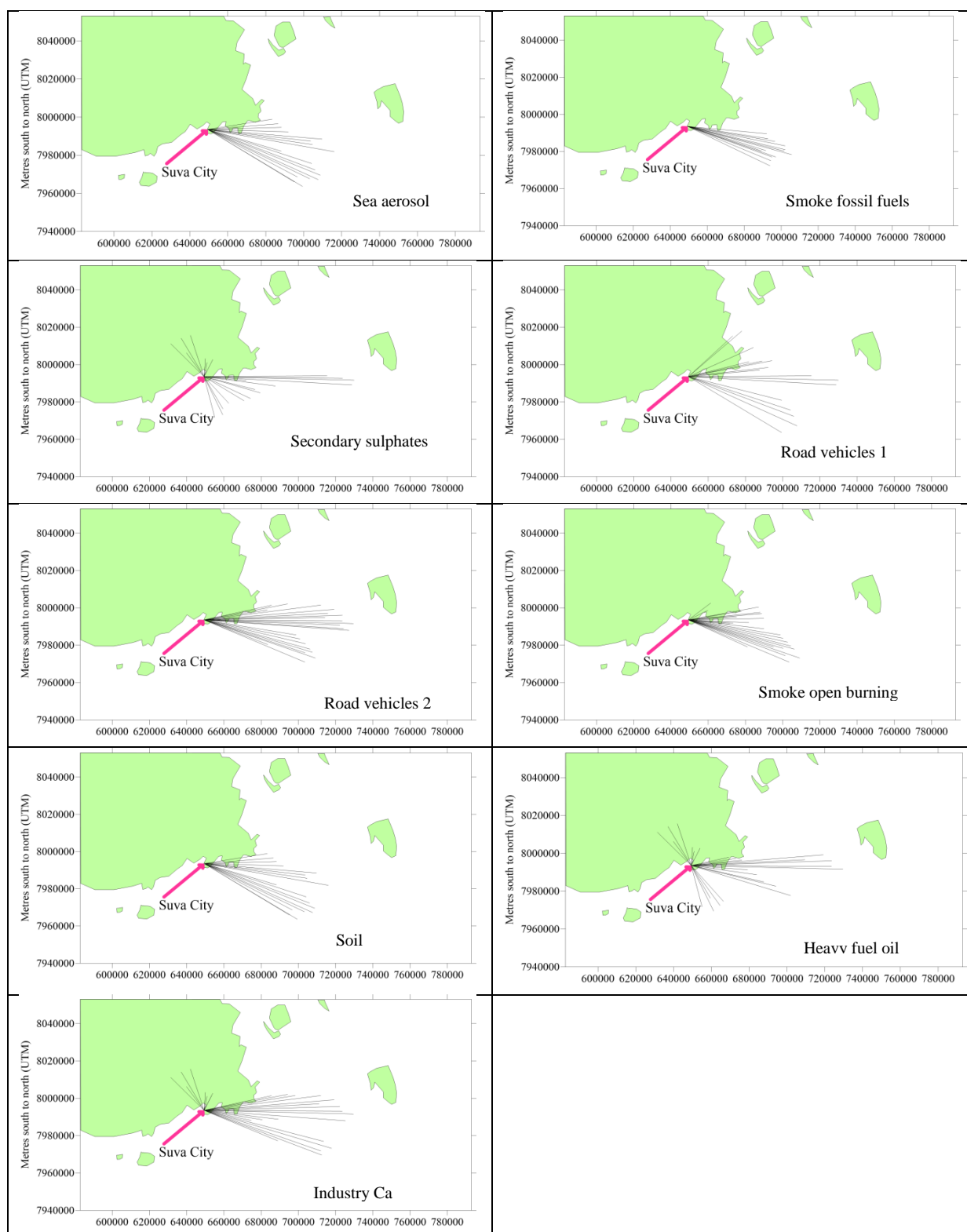


Figure A8: HYSPLIT back trajectories for peak 5% of occurrence for each source factor based on nine factor PMF model. For each source, the path of air parcels is shown for the previous hour prior to arrival in Suva, Fiji.

Section B: Sample calculation of potential reduction in mortality / mortality risk

A sample of calculations for potential mortality reductions is shown below. Whilst this example uses concentration values for road vehicle sourced PM_{2.5}, the same process was followed for each PM_{2.5} source. Table S1 also shows calculated values for road vehicle sourced PM_{2.5}. Parameters that appear both in the worked example and Table B1 are colour-coded (the same colour) to allow easy identification.

Estimated mortality benefit of reducing PM_{2.5} from road vehicles by 25%, based on PM_{2.5} mass

Baseline Mortality Rate is **Y₀ = 8 / 1000** persons per year

Exposed population is **Pop = 143,900** persons

Mortality risk (PM_{2.5} mass) is **β = 0.06** per 10 µg/m³

PM_{2.5} from road vehicles (9 factor PMF model) = 1.3 µg/m³

For 25% reduction in PM_{2.5} for road vehicle sources, Δ PM = 0.325 µg/m³

$$\begin{aligned}\Delta Y &= Y_0 (1 - e^{-\beta \Delta PM}) \text{Pop} && \text{Equation 1} \\ &= (8 / 1000) \times (1 - e^{(-0.06 / 10) \times 0.325}) \times 143,900 \\ &= 2 \text{ persons/ year (rounded to the nearest whole person)}\end{aligned}$$

Confidence interval

The confidence interval for β is 0.04 to 0.08.

Substituting these values for β into Equation 1, we now have a lower value of Δ Y = 1 and an upper value of Δ Y = 3

The calculation was repeated for results for the 8 factor PMF model, where PM_{2.5} from road vehicles = 0.8 µg/m³ (25% reduction = 0.2 µg/m³) were input into Equation 1. All other variables remained the same. This yielded Δ Y = 1 (CI 1, 2).

Combining data and uncertainties

Δ Y values for 8 and 9 factor models were averaged (to the nearest whole person) and the confidence intervals combined, to yield: Δ Y = 2 (CI 1, 3). This is the value reported in Table 4.

Risk based on PM_{2.5} components

For black carbon (BC)

β = 0.045 per 1 µg/m³ (CI 0.04, 0.07)

BC from road vehicles (9 factor PMF model) = 0.332 µg/m³

A 25% reduction would therefore be 0.083 µg/m³

Substituting these into Equation 1 gives Δ Y = 4 (CI 4, 7)

Once again, this was repeated for the 8-factor model.

This process was repeated using BC (EC) from traffic $\beta = 0.021$ per $0.26 \mu\text{g}/\text{m}^3$ (CI 0.00, 0.06) as per Thurston *et al.* (2016); yielding $\Delta Y = 8$ (CI 0, 22) for 9 factors.

For Sulphur (S)

Using $\beta = 0.14$ per $0.2 \mu\text{g}/\text{m}^3$ (CI 0.06, 0.23) as per Beelen *et al.* (2015); with $S = 0.0408$ (in road vehicle $\text{PM}_{2.5}$ for 9 factor PMF, 25% reduction = $0.01 \mu\text{g}/\text{m}^3$), Equation 1 yielded $\Delta Y = 8$ (CI 4, 13)

Table S1: Calculations for determining potential reduction in mortality for $\text{PM}_{2.5}$ due to road vehicles

	β	9 factor PMF				8 factor PMF				Source
		Concentr- ation in PM _{2.5}	10% reduction	25% reduction	100% reduction	Concentr- ation in PM _{2.5}	10% reduction	25% reduction	100% reduction	
			ΔY (persons per year)				ΔY (persons per year)			
PM_{2.5}	6% per 10 $\mu\text{g}/\text{m}^3$	1.3	1	2	9	0.8	1	1	6	Hoek <i>et al.</i> (2013)
<i>lower</i>	4% per 10 $\mu\text{g}/\text{m}^3$	1.3	1	1	6	0.8	0	1	4	
<i>upper</i>	8% per 10 $\mu\text{g}/\text{m}^3$	1.3	1	3	12	0.8	1	2	7	
BC	4.5% per 1 $\mu\text{g}/\text{m}^3$	0.332	2	4	17	0.453	2	6	23	Hoek <i>et al.</i> (2013)
<i>lower</i>	4.0% per 1 $\mu\text{g}/\text{m}^3$	0.332	2	4	15	0.453	2	5	21	
<i>upper</i>	7.0% per 1 $\mu\text{g}/\text{m}^3$	0.332	3	7	26	0.453	4	9	36	
EC from diesel (traffic)	2.1% per 0.26 $\mu\text{g}/\text{m}^3$	0.332	3	8	30	0.453	4	10	41	Thurston <i>et al.</i> (2016)
<i>lower</i>	0% per 0.26 $\mu\text{g}/\text{m}^3$	0.332	0	0	0	0.453	0	0	0	
<i>upper</i>	6% per 0.26 $\mu\text{g}/\text{m}^3$	0.332	9	22	85	0.453	12	30	114	
sulphur	14% per 0.2 $\mu\text{g}/\text{m}^3$	0.0408	3	8	32	0.0255	2	5	20	Beelen <i>et al.</i> (2015)
<i>lower</i>	6% per 0.2 $\mu\text{g}/\text{m}^3$	0.0408	1	4	14	0.0255	1	2	9	
<i>upper</i>	23% per 0.2 $\mu\text{g}/\text{m}^3$	0.0408	5	13	53	0.0255	3	8	33	

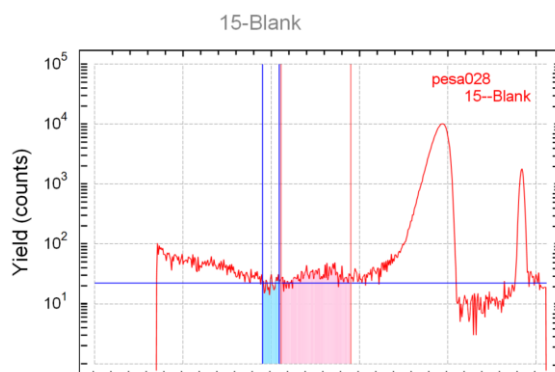
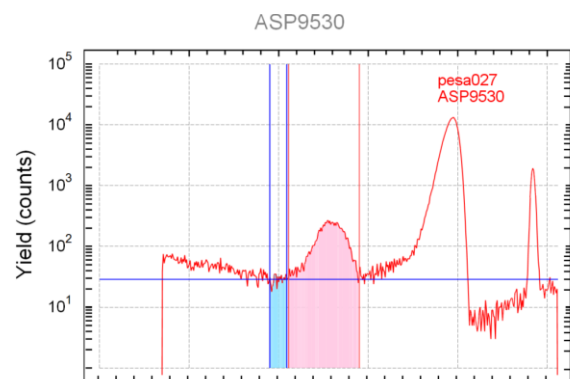
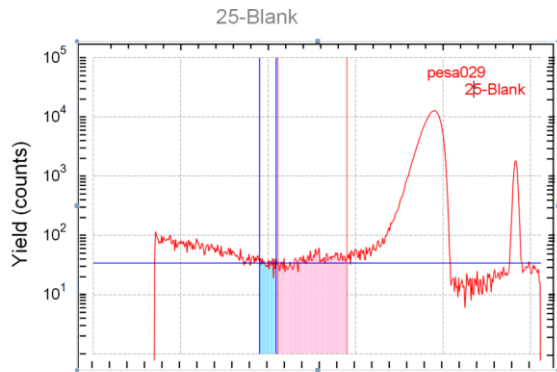
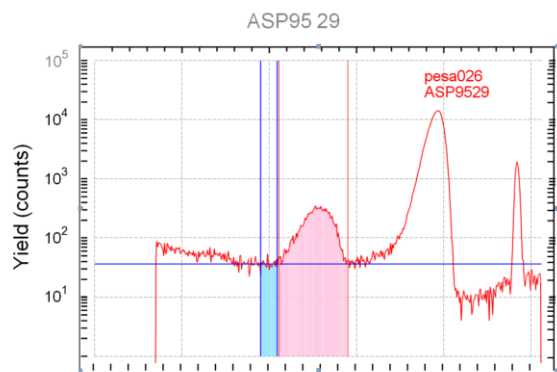
Beelen, R., Hoek, G., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z. J., Weinmayr, G., . . . Fischer, P. H., 2015. Natural-cause mortality and long-term exposure to particle components: an analysis of 19 European cohorts within the multi-center ESCAPE project. *Environmental health perspectives* 123, 525-33. 0091-6765

Hoek, G., Krishnan, R. M., Beelen, R., Peters, A., Ostro, B., Brunekreef, B., Kaufman, J. D., 2013. Long-term air pollution exposure and cardio- respiratory mortality: a review. *Environmental Health* 12, 43. 10.1186/1476-069x-12-43

Thurston, G. D., Burnett, R. T., Turner, M. C., Shi, Y., Krewski, D., Lall, R., . . . Diver, W. R., 2016. Ischemic heart disease mortality and long-term exposure to source-related components of US fine particle air pollution. *Environmental Health Perspectives* (Online) 124, 785. 1552-9924

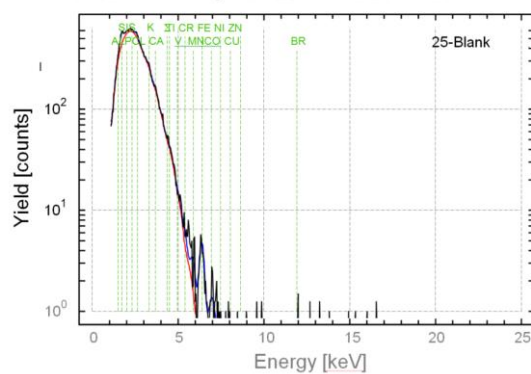
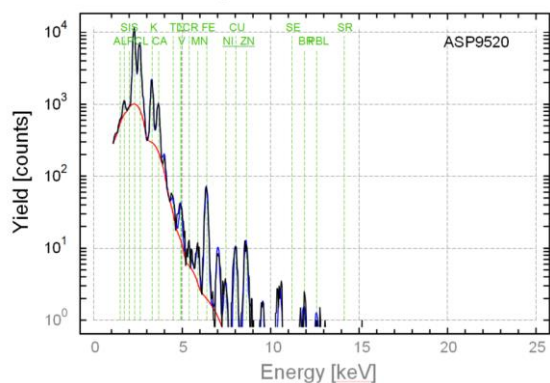
Section C: Ion beam analysis spectrum graphs and calibration

Sample of spectrum graphs for exposed filters and blanks. The upper two charts depict H determination, others are as marked.



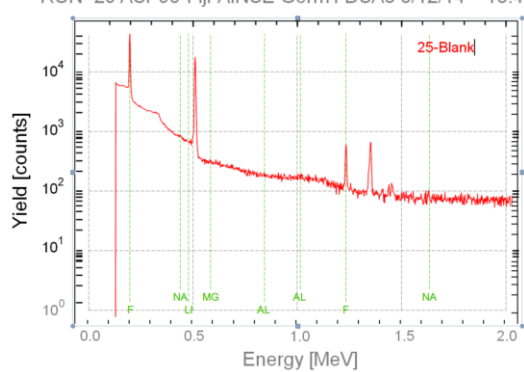
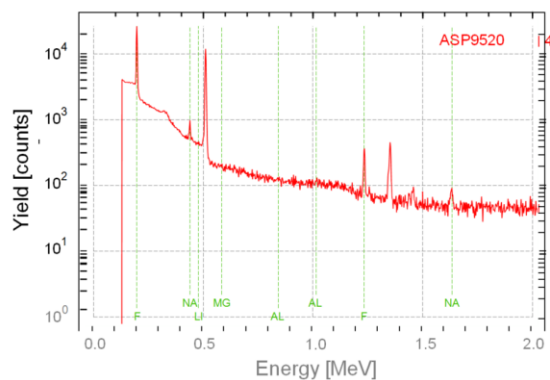
RUN=17 ASP95-Fiji-AINSE Vortex2 DS 3/12/14 12:37:12

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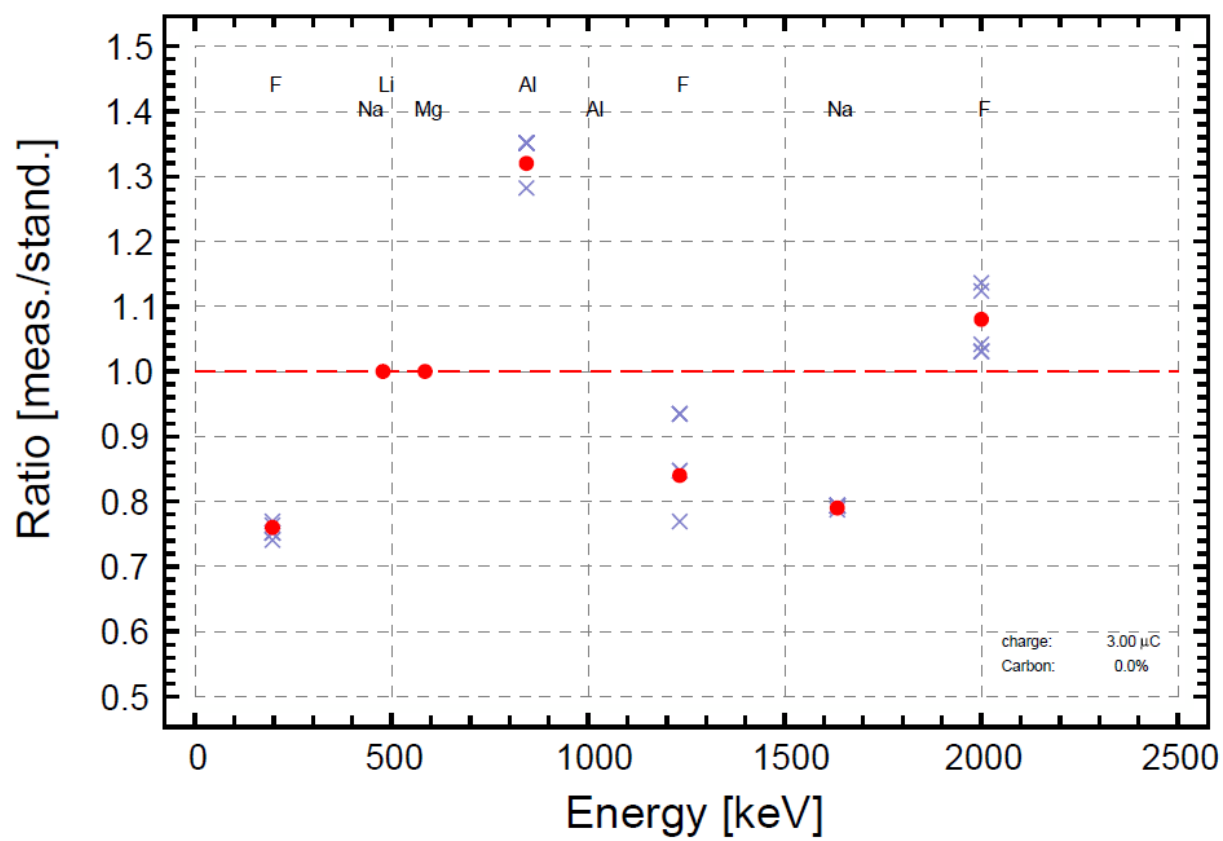


IBA calibration

RUN	BLK	SAMPLE NAME	AL	SI	CL	CA	FE	
----	-----	-----	-----	-----	-----	-----	-----	-----
		x-ray energy	1.486	1.736	2.621	3.690	6.398	
7	95907	Al-#18867	358019	0	0	18232	0	8.90477D+00
7	95907	Al-#18867	42.095	0.000	0.000	1.023	0.000	
		Standard Conc	42.900	--	--	--	--	
	Chi	1.250E+02	1.25E+02	--	--	--	--	
		calib value	0.981	--	--	--	--	
310	96210	Al-#18867	357762	0	0	18437	0	9.05931D+00
310	96210	Al-#18867	42.065	0.000	0.000	1.035	0.000	
		Standard Conc	42.900	--	--	--	--	
	Chi	1.345E+02	1.34E+02	--	--	--	--	
		calib value	0.981	--	--	--	--	
8	95908	SiO-#18876	0	504244	0	17180	0	7.93364D+00
8	95908	SiO-#18876	0.000	28.510	0.000	0.964	0.000	
		Standard Conc	--	28.310	--	--	--	
	Chi	2.268E+01	--	2.27E+01	--	--	--	
		calib value	--	1.007	--	--	--	
311	96211	SiO-#18876	0	505939	0	17234	0	7.95906D+00
311	96211	SiO-#18876	0.000	28.605	0.000	0.967	0.000	
		Standard Conc	--	28.310	--	--	--	
	Chi	4.933E+01	--	4.93E+01	--	--	--	
		calib value	--	1.010	--	--	--	
9	95909	NaCl-#37118	0	0	765918	18103	0	6.33196D+00
9	95909	NaCl-#37118	0.000	0.000	29.593	1.016	0.000	
		Standard Conc	--	--	30.510	--	--	
	Chi	6.488E+02	--	--	6.49E+02	--	--	
		calib value	--	--	0.970	--	--	
312	96212	NaCl-#37118	0	0	766465	18358	0	6.13379D+00
312	96212	NaCl-#37118	0.000	0.000	29.614	1.030	0.000	
		Standard Conc	--	--	30.510	--	--	
	Chi	6.185E+02	--	--	6.19E+02	--	--	
		calib value	--	--	0.971	--	--	
10	95910	CaF2-#37134	0	5927	0	462222	0	1.04048D+01
10	95910	CaF2-#37134	0.000	0.335	0.000	25.944	0.000	
		Standard Conc	--	--	--	26.280	--	
	Chi	7.422E+01	--	--	--	7.42E+01	--	
		calib value	--	--	--	0.987	--	
313	96213	CaF2-#37134	0	5462	0	462149	0	1.00229D+01
313	96213	CaF2-#37134	0.000	0.309	0.000	25.940	0.000	
		Standard Conc	--	--	--	26.280	--	
	Chi	7.600E+01	--	--	--	7.60E+01	--	
		calib value	--	--	--	0.987	--	
11	95911	Fe-#16572	0	0	0	18390	225035	1.45775D+01
11	95911	Fe-#16572	0.000	0.000	0.000	1.032	48.429	
		Standard Conc	--	--	--	--	47.800	
	Chi	3.729E+01	--	--	--	--	3.73E+01	
		calib value	--	--	--	--	1.013	
314	96214	Fe-#16572	0	0	0	18415	224645	1.46705D+01
314	96214	Fe-#16572	0.000	0.000	0.000	1.034	48.345	
		Standard Conc	--	--	--	--	47.800	
RUN	BLK	SAMPLE NAME	AL	SI	CL	CA	FE	
----	-----	-----	-----	-----	-----	-----	-----	-----
		x-ray energy	1.486	1.736	2.621	3.690	6.398	
	Chi	2.800E+01	--	--	--	--	2.80E+01	
		calib value	--	--	--	--	1.011	
	Chi	3.631E+02	1.30E+02	3.60E+01	6.34E+02	7.51E+01	3.26E+01	
total calib			0.9919	0.9809	1.0087	0.9703	0.9871	1.0123
STDV total			0.0003	0.0017	0.0003	0.0001	0.0009	

RUN	BLK		SAMPLE	F (AREA) ug/cm2	F (197) ug/cm2	Na (440) ug/cm2	Li (478) ug/cm2	Mg (585) ug/cm2	Al (843) ug/cm2	Al (1014) ug/cm2	F (1233) ug/cm2	Na (1634) ug/cm2
		=====	=====	=====	=====	=====	=====	=====	=====	=====	=====	=====
		CONCFACOR	original	217.2700	155.3000	20.6000	0.7291	0.0026	5.1500	1.4000	1.9500	14.9500
7	35907	ptanal area	Al-#18867	1423	283	222	---	---	854	605	---	75
7	35907	peak energy	Al-#18867		198	441	---	---	844	1014	---	1632
		nom. conc.	Al-#18867	---	---	---	---	---	42.900	42.900	---	---
		ug/cm2 wos	Al-#18867						55.301	144.121		
		CONCFACOR		217.27	155.30	20.60	0.73	0.00	6.64	4.70	1.95	14.95
		CONCFACOR	org/new	0.00	0.00	0.00	0.00	0.00	0.78	0.30	0.00	0.00
310	36210	ptanal area	Al-#18867	1237	219	212	---	---	900	564	---	65
310	36210	peak energy	Al-#18867		197	441	---	---	844	1014	---	1632
		nom. conc.	Al-#18867	---	---	---	---	---	42.900	42.900	---	---
		ug/cm2 wos	Al-#18867						55.223	134.321		
		CONCFACOR		217.27	155.30	20.60	0.73	0.00	6.99	4.38	1.95	14.95
		CONCFACOR	org/new	0.00	0.00	0.00	0.00	0.00	0.74	0.32	0.00	0.00
8	35908	ptanal area	SiO-#18876	1256	239	209	---	---	---	---	---	65
8	35908	peak energy	SiO-#18876		198	441	---	---	---	---	---	1634
		nom. conc.	SiO-#18876	---	---	---	---	---	---	---	---	---
		ug/cm2 wos	SiO-#18876									
		CONCFACOR		217.27	155.30	20.60	0.73	0.00	6.99	4.38	1.95	14.95
		CONCFACOR	org/new	0.00	0.00	0.00	0.00	0.00	0.74	0.32	0.00	0.00
311	36211	ptanal area	SiO-#18876	1072	210	219	---	---	---	---	---	62
311	36211	peak energy	SiO-#18876		197	441	---	---	---	---	---	1633
		nom. conc.	SiO-#18876	---	---	---	---	---	---	---	---	---
		ug/cm2 wos	SiO-#18876									
		CONCFACOR		217.27	155.30	20.60	0.73	0.00	6.99	4.38	1.95	14.95
		CONCFACOR	org/new	0.00	0.00	0.00	0.00	0.00	0.74	0.32	0.00	0.00
9	35909	ptanal area	NaCl-#37118	1213	240	4438	---	---	---	---	---	699
9	35909	peak energy	NaCl-#37118		198	441	---	---	---	---	---	1632
		nom. conc.	NaCl-#37118	---	---	19.790	---	---	---	---	---	19.790
		ug/cm2 wos	NaCl-#37118			71.806						15.592
		CONCFACOR		217.27	155.30	74.74	0.73	0.00	6.99	4.38	1.95	11.78
		CONCFACOR	org/new	0.00	0.00	0.28	0.00	0.00	0.74	0.32	0.00	1.27
312	36212	ptanal area	NaCl-#37118	1264	285	4990	---	---	---	---	---	703
312	36212	peak energy	NaCl-#37118		197	441	---	---	---	---	---	1633
		nom. conc.	NaCl-#37118	---	---	19.790	---	---	---	---	---	19.790
		ug/cm2 wos	NaCl-#37118			80.748						15.677
		CONCFACOR		217.27	155.30	84.05	0.73	0.00	6.99	4.38	1.95	11.84
		CONCFACOR	org/new	0.00	0.00	0.25	0.00	0.00	0.74	0.32	0.00	1.26
10	35910	ptanal area	CaF2-#37134	16994	8614	263	---	---	---	---	112	72
10	35910	peak energy	CaF2-#37134		198	441	---	---	---	---	1236	1634
		nom. conc.	CaF2-#37134	24.920	24.920	---	---	---	---	---	24.920	---

PIGE Calibration



Appendix D:

Supplementary Information for Paper Five

Radiocarbon determination of fossil and modern carbon contribution to aerosol in Suva, Fiji

C. F. Isley, P. F. Nelson, M.P. Taylor, A. Alan Williams and Geraldine Jacobsen

Section A. Tables and Figures

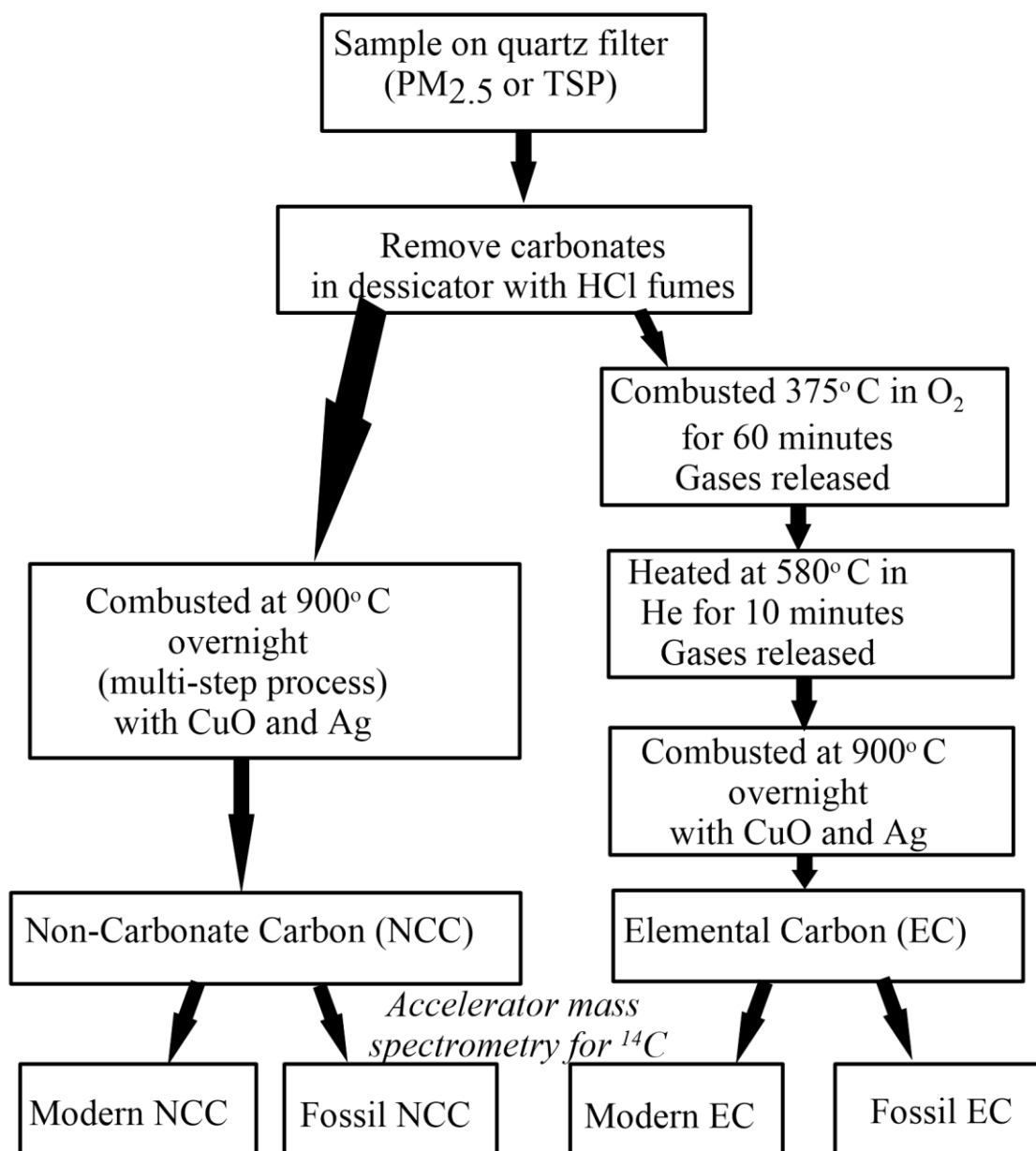


Fig. S1. Sample preparation method. This uses the method of (Szidat *et al.* 2013). For the combustion at 900°C, samples were heated to 900°C and held for 2 hours at that temperature. Samples were then cooled to 600°C and held for 2 hours before cooling to room temperature.

Table S1. Sampling schedule. Sample names identify the location and type of sample, in terms of air sample particle fraction.

Sample	Air volume (m ³)	Sample Use	Date sampled	Notes (Table 1). Air samples were composites of multiple samples collected at each site. The sample collection dates are given along with the cumulative air volume represented by each composite air sample. Samples with sufficient estimated carbon content were split and used for determination of both total non-carbonate carbon (total
City_PM _{2.5} _1	90	Total NCC + EC/OC	27-28/4/15, 9/5/15	
City_PM _{2.5} _2	90	Total NCC + EC/OC	11-12/5/15, 14/5/15	
City_PM _{2.5} _3	60	Total NCC + EC/OC	23-24/10/15	
City_PM _{2.5} _4	60	Total NCC only, rest destroyed	25-26/10/15	
City_PM _{2.5} _5	60	Total NCC only, rest destroyed	15-16/5/2015	
City_TSP_1	20	Total NCC + EC/OC	27/10/14 to 31/10/14	
City_TSP_2	29	Total NCC only, rest destroyed	21/10/14 to 26/10/14	
City_TSP_3	29	Total NCC + EC/OC	29-31/10/14, 10-17/5/15	
Kinoya_TSP_1	20	Total NCC only	27-31/10/14	
Kinoya_TSP_2	82	Total NCC + EC/OC	20/4/15-10/5/15	
Kinoya_TSP_3 (duplicate)	57	Total NCC + EC/OC	20-26/10/15	
Kinoya_TSP_4 (duplicate)	51	Total NCC + EC/OC	20-26/10/15	
Suva Point_TSP_1	103	Total NCC only	21-31/10/14, 23-30/4/15, 8-11/5/15	
Suva Point_TSP_2	181	Total NCC only	30/4-8/5/15, 15/5/15-9/6/15	
Suva Point_TSP_3 (duplicate)	181	Sample destroyed	30/4-8/5/15, 15/5/15-9/6/15	

NCC) and separately for determining elemental carbon (EC) components. Three half-samples and one full sample were destroyed due to furnace problems, as listed. The proportion of the filter and hence volume used for NCC and EC analysis are not always 50%. This is particularly the case where a portion of sample was destroyed. The filter proportion for each fraction was determined by mass.

Table S2. Concurrent data available measurement of TSP, PM_{2.5} and black carbon at all sites (Isley *et al.* 2017a; Isley *et al.* 2018)

Parameter	Kinoya	Suva Point	Suva City
PM _{2.5} concentration μg m ⁻³ (Isley <i>et al.</i> 2017a), Osiris	11.8 ± 0.1	3.8 ± 0.2	9.7 ± 0.1
TSP concentration μg m ⁻³ (Isley <i>et al.</i> 2017a) Osiris	44.2 ± 0.4	9.1 ± 0.1	28.4 ± 0.2
Ultrafine particle number concentration cm ⁻³ (Isley <i>et al.</i> 2017b)	4.4 ± 0.2 × 10 ⁵	1.48 ± 0.04 × 10 ⁵	16.4 ± 0.2 × 10 ⁵
Black carbon concentration μg m ⁻³ (Isley <i>et al.</i> 2017a; Isley <i>et al.</i> 2017b)	1.4 ± 0.2	0.2 ± 0.2	2.2 ± 0.1

Table S3. Non-carbonate carbon (NCC) results, corrected for blanks. Contemporary carbon is corrected for leaf samples. Ranges shown for mean contemporary carbon values include standard error and uncertainty stemming from the unknown age of plant materials burned.

Sample	Ambient total NCC concentration ($\mu\text{g m}^{-3}$ in air)	Contemporary carbon % of total NCC
City_PM _{2.5} _1	5.9	23
City_PM _{2.5} _2	4.2	20
City_PM _{2.5} _3	5.9	19
City_PM _{2.5} _4	6.0	21
City_PM _{2.5} _5	5.0	14
<i>Mean City PM_{2.5}</i>	<i>5.4 ± 0.3</i>	<i>19 (17 - 21)</i>
City_TSP_1	16	13
City_TSP_2	9	22
City_TSP_3	13	22
<i>Mean City TSP</i>	<i>13 ± 2</i>	<i>19 (16 - 22)</i>
<i>Mean City -all</i>		<i>19 (17 - 21)</i>
Kinoya_TSP_1	11	43
Kinoya_TSP_2	11	54
Kinoya_TSP_3	15	47
Kinoya_TSP_4	15	46
<i>Mean Kinoya</i>	<i>13 ± 1</i>	<i>48 (46 – 50)</i>
Suva Point_TSP_1	3.1	23
Suva Point_TSP_2	2.3	34
<i>Mean Suva Point</i>	<i>2.7 ± 0.4</i>	<i>29 (23 – 35)</i>

Table S4. Elemental carbon (EC) results, corrected for blanks. Standard error is shown. Contemporary carbon is corrected for leaf samples. Ranges shown for mean contemporary carbon values include standard error and uncertainty stemming from the unknown age of plant materials burned as well as error from uncertainty in assignment of the EC fraction.

Sample	Ambient total EC concentration ($\mu\text{g m}^{-3}$ in air)	Contemporary carbon % of total EC	Percentage of EC in NCC
City_PM _{2.5} _1	1.2	1.1	19
City_PM _{2.5} _2	1.6	2.4	37
City_PM _{2.5} _3	1.9	2.4	31
<i>Mean City PM_{2.5}</i>	<i>1.5 ± 0.5</i>	<i>1.9 (1.5– 2.3)</i>	<i>28 ± 10</i>
City_TSP_1	3.06	4	19
City_TSP_3	2.91	8	23
<i>Mean City TSP</i>	<i>3 ± 1</i>	<i>6 (4 – 8)</i>	<i>21 ± 10</i>
Kinoya_TSP_2	2.2	20	18
Kinoya_TSP_3	2.4	11	15
Kinoya_TSP_4	2.6	17	17
<i>Mean Kinoya</i>	<i>2.4 ± 1</i>	<i>16 (14 – 18)</i>	<i>18 ± 10</i>

Table S5: Organic carbon (OC) ambient mass concentration results were determined using $OC = NCC - EC$.
 . Contemporary carbon percentages were calculated by:

- (1) Determining the mass of contemporary NCC and EC in each sample, using ambient NCC and EC concentration multiplied by percentage contemporary from Table S3 and S4.
- (2) Determining the mass of contemporary OC, using
 $OC_{\text{contemporary}} = NCC_{\text{contemporary}} - EC_{\text{contemporary}}$ for mass.
- (3) Calculating the percentage of OC mass represented by this contemporary fraction.
- (4) Using NCC data for the uncertainty stemming from the unknown age of plant materials burned (Table S3) and EC data for uncertainty in EC/OC fraction split, ranges were shown to indicate the uncertainty of the mean contemporary carbon percentage for each site.

Sample	Ambient total OC concentration ($\mu\text{g m}^{-3}$ in air) Standard error is shown	Contemporary carbon % of total OC	Percentage of OC in NCC
City_PM _{2.5} _1	4.8	28	81
City_PM _{2.5} _2	2.7	30	63
City_PM _{2.5} _3	4.1	27	69
Mean City PM_{2.5}	3.8 ± 0.6	26 (23 – 28)	72 ± 10
City_TSP_1	13	16	71
City_TSP_3	10	27	81
Mean City TSP	12 ± 1	19 (16 – 21)	77 ± 10
Kinoya_TSP_2	9.8	62	82
Kinoya_TSP_3	13.2	53	86
Kinoya_TSP_4	12.3	52	81
Mean Kinoya	12 ± 1	55 (53 - 57)	82 ± 10

Table S6. OC/EC and EC/TC ratios

	OC/EC in total NCC*	Contemporary OC/ Contemporary EC	Fossil OC/ Fossil EC	EC/TC in total NCC	Contemporary EC/TC	Fossil EC/TC
City PM _{2.5}	2.5	34	2	0.28	0.34	0.03
City TSP	4	13	3.5	0.20	0.22	0.07
Kinoya TSP	5	17	2.6	0.17	0.27	0.06

* These OC/EC ratios are averaged over all samples. The primary OC/EC ratio for the Suva airshed (OC/EC)_p, was 1.69 for PM_{2.5} and 2.5 for TSP, being the minimum OC/EC ratio over all samples

Table S7. Sensitivity analysis of sources. All values are concentration in air ($\mu\text{g}/\text{m}^3$)

Scenario	EC	EC fossil	OC	OC fossil	OC contemporary	EC biomass	OC primary fossil	OC biomass	OC biogenic	OC fossil secondary
city pm2.5	1.5	1.5	3.9	2.9	1.0	0.03	0.9	0.18	0.8	2.0
city TSP	3.0	2.8	12.0	9.7	2.3	0.18	1.6	1.08	1.2	8.1
Kinoya TSP	2.4	2.0	2.4	5.3	6.5	0.38	1.2	2.29	4.2	4.1
City PM_{2.5}										
Low EC	1.0	1.0	4.4	3.1	1.2	0.02	0.6	0.12	1.1	2.5
high EC	2.0	2.0	3.4	2.4	0.9	0.04	1.2	0.23	0.7	1.3
low contemporary	1.5	1.5	3.9	3.0	0.9	0.02	0.9	0.14	0.8	2.1
OCfossil_P	1.5	1.5	3.9	2.9	1.0	0.03	0.7	0.18	0.8	2.1
low OCfossil_P	1.5	1.5	3.9	2.9	1.0	0.03	1.8	0.18	0.8	1.1
high low	1.5	1.5	3.9	2.9	1.0	0.03	0.9	0.03	1.0	2.0
OCbiomass/ECbiomass	1.5	1.5	3.9	2.9	1.0	0.03	0.9	0.44	0.6	2.0
High OCbiomass/ECbiomass	1.5	1.5	3.9	2.9	1.0	0.03	0.9	0.44	0.6	2.0
min						0.02	0.6	0.03	0.6	1.1
max						0.04	1.8	0.44	1.1	2.5
City TSP										
Low EC	2.0	1.9	13.0	10.2	2.8	0.12	1.1	0.72	2.0	9.2
high EC	4.0	3.7	11.0	8.7	2.3	0.24	2.2	1.44	0.9	6.5
low contemporary	3.0	2.6	11.2	9.4	1.8	0.12	1.5	0.72	1.1	7.9
OCfossil_P	3.0	2.8	12.0	9.7	2.3	0.18	1.3	1.08	1.2	8.4
low OCfossil_P	3.0	2.8	12.0	9.7	2.3	0.18	3.4	1.08	1.2	6.3
high low	3.0	2.8	12.0	9.7	2.3	0.18	1.6	0.18	2.1	8.1
OCbiomass/ECbiomass	3.0	2.8	12.0	9.7	2.3	0.18	1.6	2.69		8.1
High OCbiomass/ECbiomass	3.0	2.8	12.0	9.7	2.3	0.18	1.6	2.69		8.1
min						0.12	1.1	0.18	0.9	6.3
max						0.24	3.4	2.69	2.1	9.2
Kinoya TSP										
Low EC	1.4	1.2	12.8	5.5	7.3	0.22	0.7	1.33	6.0	4.8
high EC	3.4	2.8	10.8	4.6	6.2	0.54	1.6	3.25	2.9	3.0
low contemporary	2.4	2.1	11.8	5.5	6.3	0.33	1.2	2.29	4.5	3.9
OCfossil_P	2.4	2.0	2.4	5.3	6.5	0.38	1.0	2.29	4.2	4.3
low OCfossil_P	2.4	2.0	2.4	5.3	6.5	0.38	2.4	2.29	4.2	2.9
high low	2.4	2.0	2.4	5.3	6.5	0.38	1.2	0.38	6.1	4.1
OCbiomass/ECbiomass	2.4	2.0	2.4	5.3	6.5	0.38	1.2	5.73	0.8	4.1
High OCbiomass/ECbiomass	2.4	2.0	2.4	5.3	6.5	0.38	1.2	5.73	0.8	4.1
min						0.22	0.7	0.38	0.8	2.9
max						0.54	2.4	5.73	6.1	4.8

Section B. Analysis of leaf samples

Plant species for leaf samples were identified using Thaman (2012); the Fijian and botanical names of each plant are listed here:

- Two leaf samples were collected from Suva City;
 - *Kava* (*Piper methysticum*) was collected from the roadside between the markets and port area and
 - *Sacasaca* (*Codiaeum variegatum*) from outside the Suva cinema complex.
- *Uto* (*Artocarpus altilis*) was collected from the residential area of Kinoya, near Velau Drive.
- *Senitoa* (*Hibiscus rosa sinensis*) was collected from the University of the South Pacific lower campus at Suva Point.
- *Bui ni pusi* (*Acalypha hispida*) was collected from Colo-i-Suva, a forest area in the northeast of Suva.

In order to ensure that leaf samples did not contain surface contaminants, a cleaning process was carried out for each leaf sample. A leaf segment 5 cm long was placed in a 60°C heat bath for 50 minutes in a 2M HCl solution, rinsed with MilliQ water, returned to the heat bath for 50 minutes with 4 % NaOH solution, rinsed with Milli-Q water, placed in a 60°C heat bath again for 50 minutes in a 2M HCl solution (to remove any atmospheric derived carbonate in the NaOH), then rinsed three times with MilliQ water. After 18 hours in the drying oven at 60°C, samples were placed in glass vials. Each leaf sample was weighed into a pre-cleaned quartz combustion tube and analysed as per NCC samples.

References.

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- Isley, C. F., Nelson, P. F., Taylor, M. P., Mazaheri, M., Morawska, L., 2017b. Airborne ultrafine particles in a Pacific Island Country: characteristics, sources and implication on human exposure *Environmental Pollution* 231, 367-378.
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Appendix E:

Supplementary Information for Paper Six

Ambient air quality and indoor exposure:

PM_{2.5} implications for health in Suva.

C. F. Isley, P. F. Nelson, M.P. Taylor, A. L. Morrison,

A. J. Atanacio, E. Stelcer and D. D. Cohen

Section A: Tables and Figures

Table S1: PM_{2.5} and BC concentrations for Microvol sampling locations, Suva, Fiji. Also shown are concurrent PM_{2.5} results from the nearest ambient monitoring site and concurrent gravimetric PM_{2.5} results from Suva City.

Site	Microvol PM _{2.5}	Microvol BC	Distance from ambient monitoring site	Nearest ambient PM _{2.5} measurement (Photometer)	Gravimetric PM _{2.5} - City (ASP)	ASP BC
Indoor locations						
Toorak 1	10.7 ± 1.1	1.6 ± 0.4	850 m W City	8.1 ± 0.1	6.1 ± 0.2	2.5 ± 1.0
Toorak 2	10.5 ± 1.6	1.5 ± 0.2	600 m W City	3.5 ± 0.1	7.5 ± 0.7	2.7 ± 0.3
Caubati	19.8 ± 0.7	1.1 ± 0.2	550 m NW Kinoya	8.3 ± 0.2	6.4 ± 0.2	1.3 ± 0.1
Kinoya	12.7 ± 1.5	1.4 ± 0.2	30 m Kinoya Indoors	10.4 ± 0.2	8.7 ± 0.7	3.1 ± 0.4
Namadi Heights	5.8 ± 0.3	1.0 ± 0.1	2.8 km SW Kinoya 3.5 km NE City	12.7 ± 0.2 9.1 ± 0.1	6.8 ± 0.5	2.7 ± 0.5
Navua	10.5 ± 0.1	2.1 ± 0.1	28 km SW City	10.6 ± 0.1	8.9 ± 2.2	2.1 ± 0.9
Wainbuku	7.6 ± 0.5	1.0 ± 0.2	7.2 km NE Kinoya	13.2 ± 0.3	7.7 ± 0.9	3.4 ± 0.6
Flagstaff 1	7.7 ± 0.5	1.6 ± 0.1	1.8 km SE City	8.3 ± 0.1	6.2 ± 0.2	2.3 ± 0.9
Flagstaff 2	7.8 ± 0.5	1.7 ± 0.1	1.7 km SE City	10.1 ± 0.1	15.4 ± 2.4	3.5 ± 0.5
Outdoor locations						
Suva Point	5.5 ± 0.8	0.2 ± 0.2	5 m Suva Point	8.3 ± 0.2	7.4 ± 0.8	3.7 ± 0.6
City	8.6 ± 0.5	2.9 ± 0.1	10 m City	equipment malfunction	9.0 ± 0.9	2.1 ± 0.8
Walu Bay	10.8 ± 0.6	3.5 ± 0.3	1.2 km NE City	equipment malfunction	6.7 ± 0.7	2.2 ± 0.8

PM_{2.5} source factors in Suva city centre, Fiji

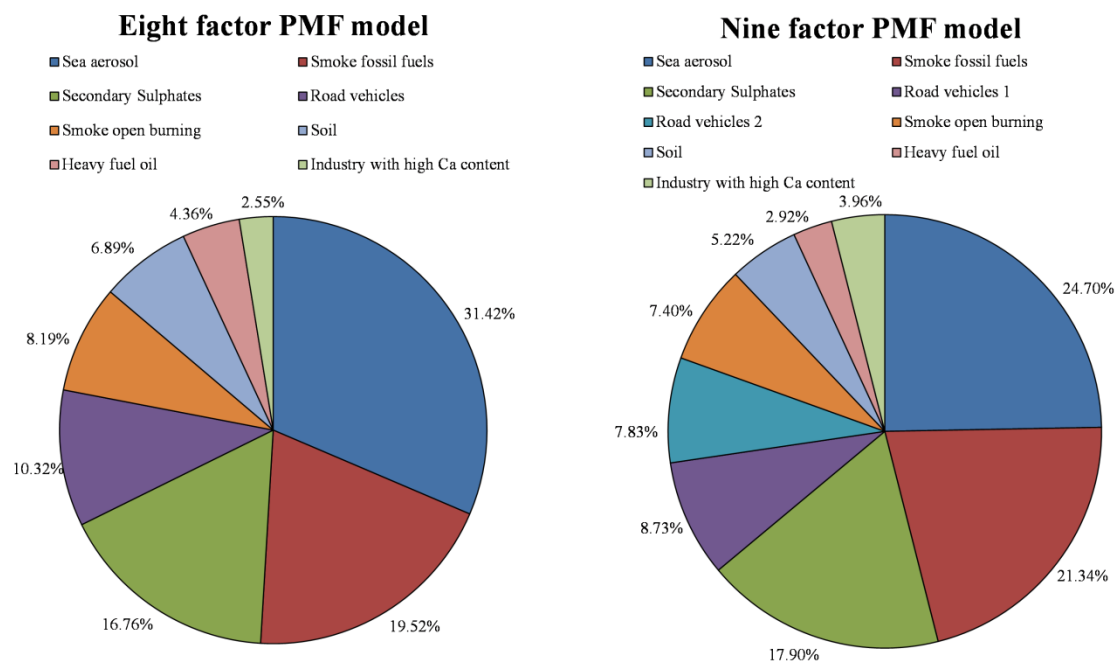


Figure S1: PM_{2.5} source factors for Suva City, 2014-2015. Data from Isley *et al.* (2018)

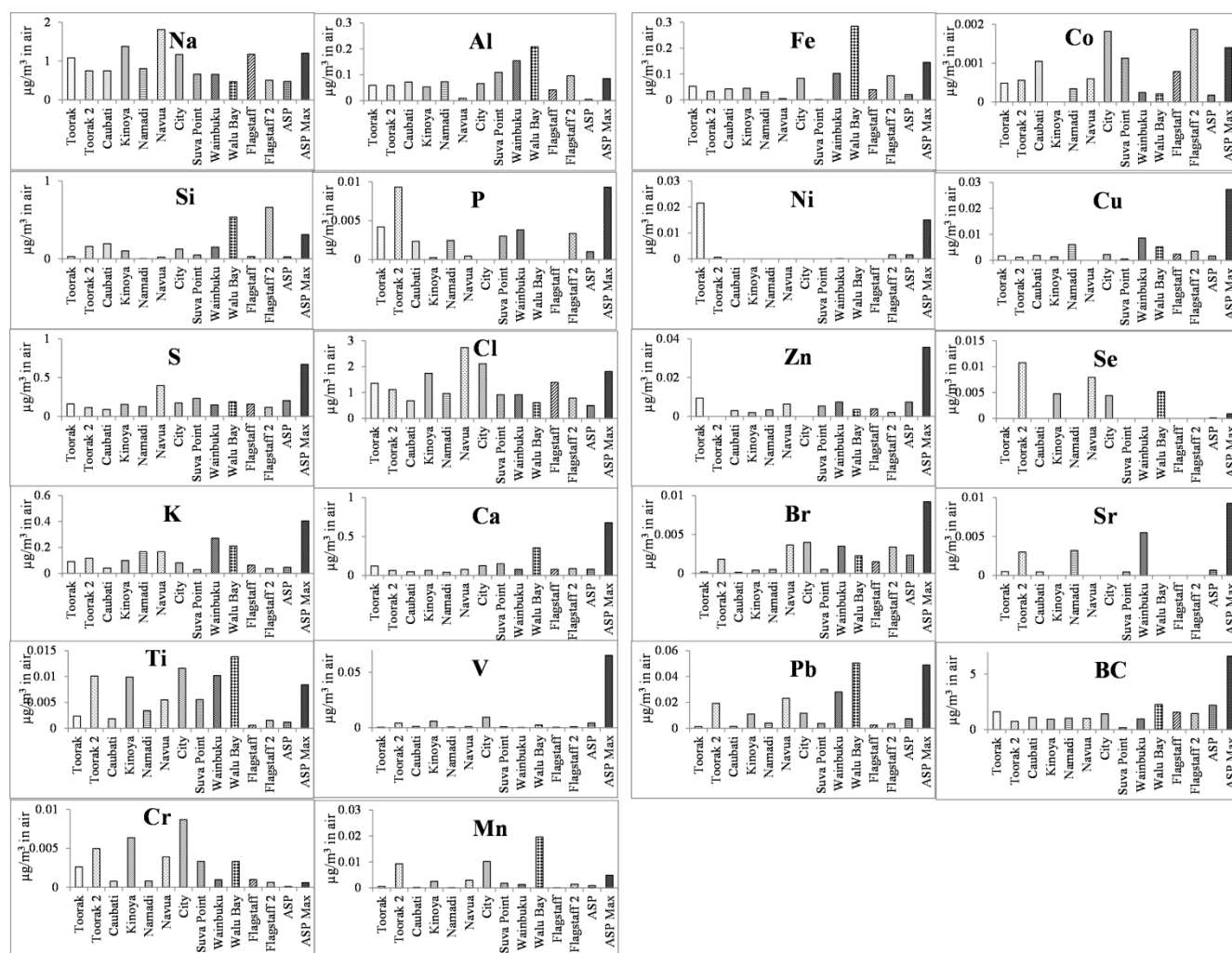


Figure S2: Elemental concentration data for each element, average over all Microvol samples for each site. ASP data included here is the annual mean of all samples collected at Suva City from October 2014 to October 2015. Maximum concentration recorded in ASP samples is also given in order to demonstrate the variation in elemental concentration recorded at the city site over a year.

Section B: Photographs

Indoor sampling locations



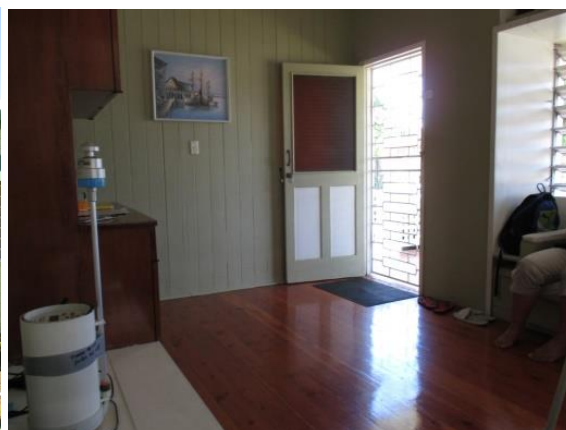
Toorak 1: View from sampling site showing Walu Bay industries



Caubati: The barracks



Toorak 2. The back windows (right) had only lattice and no close-able covering, allowing constant air exchange with outside.



Flagstaff 1

Indoor sampling locations (continued)



The road at Flagstaff (outside Flagstaff 2)

Outdoor sampling locations



Suva City. An ASP sampler (green) is affixed to the tower that also houses Australian Bureau of Meteorology instruments. An Osiris sampler (white) is affixed on poles adjacent to the tower.

Outdoor sampling locations (continued)



The Walu Bay sampling site was surrounded by large and small industrial premises



The Suva Point sampler was affixed to poles extending from the upper veranda of the jetty lab (yellow roof) on the lower campus of the University of the South Pacific. This photo demonstrates the ocean-exposure of this site.



View from the Kinoya sampler (left). In the street near the Kinoya sampler (right), there is frequently waste-burning.

Fijian houses



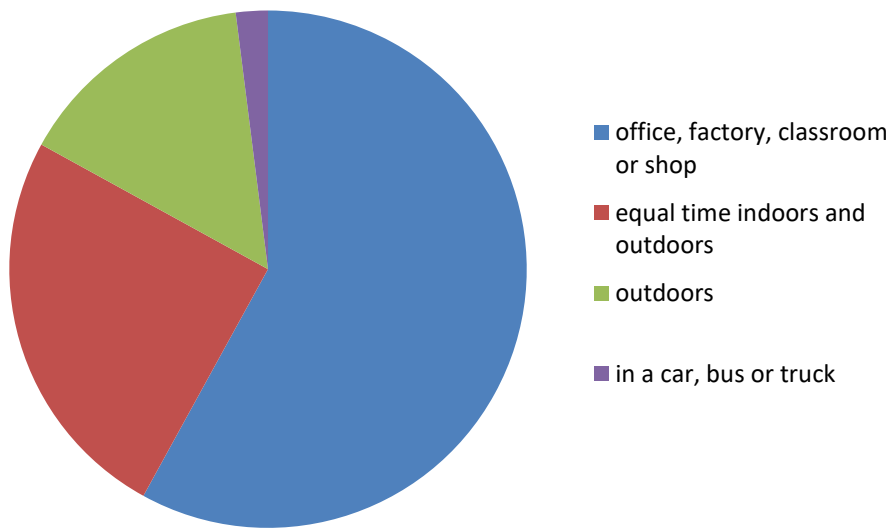
These houses around Suva (2016) near Kinoya (top left), Flagstaff (top right) and Lami (bottom) demonstrate that many Fijian homes are built to allow airflow. This means that indoor air quality is more strongly affected by outside air emissions. The concrete plant at Lami is visible in the background (bottom right).

Supplementary Section C: Survey questions and responses

Survey results represent 25 different neighbourhoods within Greater Suva, including 81 households from within the Suva City Council area. Demographic questions (including age, residential suburb and household size) were included to ensure a relevant population sample; questions and survey format were developed as part of the ethics approval for this project.

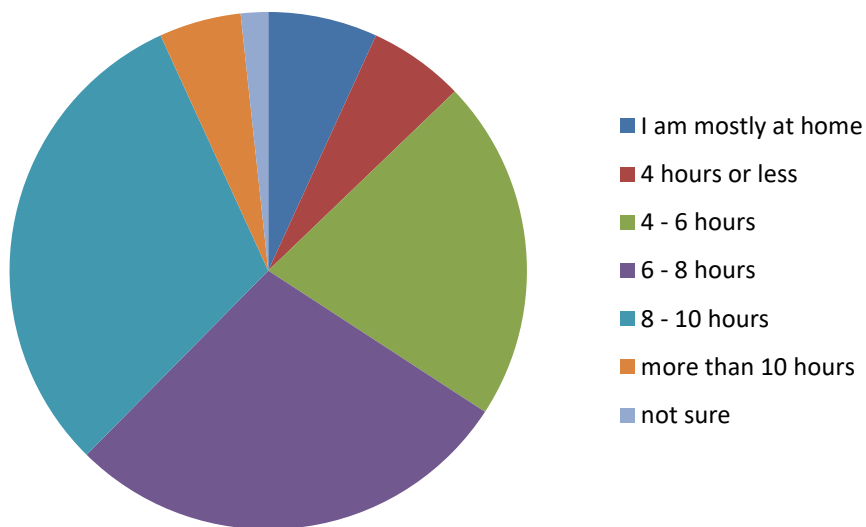
Questions relating to this paper:

1. Where do you spend most of your time during working hours?
 - In an office, factory, workshop, classroom or shop
 - In a car, bus or truck
 - Inside a house
 - Outdoors
 - Equal time indoors and outdoors
 - Other



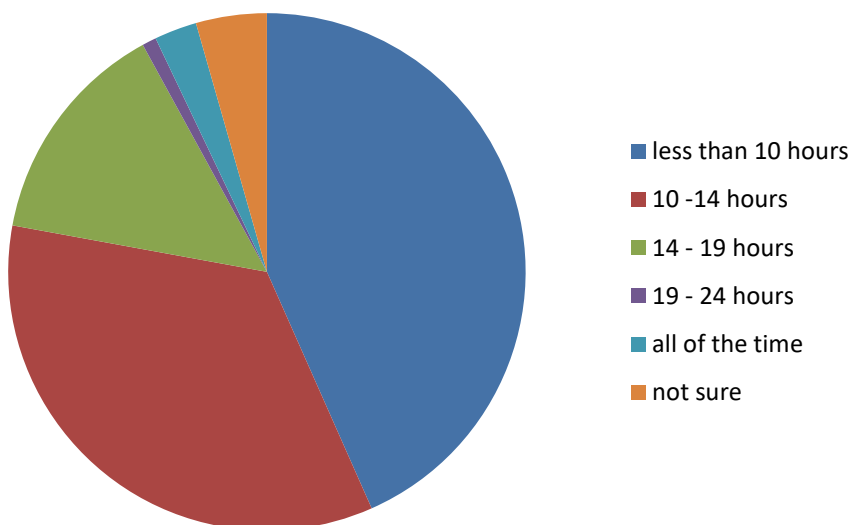
2. How long do you spend at your place of work or study each day?

- I am mostly at home
- 4 hours or less
- 4-6 hours
- 6-8 hours
- 8-10 hours
- More than 10 hours
- Not sure



3. How much time would you spend indoors at home on a normal work day (include sleeping time)?

- Less than 10 hours (eg. from 10PM to 8AM)
- 10-14 hours (eg. from 6PM to 8AM)
- 14 - 19 hours (eg. from 3PM to 10AM)
- 19 - 24 hours (most of the time)
- all of the time
- not sure



Isley, C. F., Nelson, P. F., Taylor, M. P., Stelcer, E., Atanacio, A. J., Cohen, D. D., . . . Maata, M., 2018. Reducing mortality risk by targeting specific air pollution sources: Suva, Fiji. *Sci Total Environ* 612C 450-461.

Appendix F:

Supplementary Information for Paper Seven

Air quality management in the Pacific Islands: a review of past performance and implications for future directions

C.F. Isley and M. P. Taylor

All photos were taken by C. F. Isley from
November 2013 to November 2016

Section A: Pictures of waste-burning in Suva Fiji



A waste pile in a suburban yard near Valelevu shows evidence of burning paper, textiles, a mattress, plastics and a LPG gas bottle (2014).



Smoke from burning waste hangs over the inner city suburbs (Suva 2015).



Waste pile burning in a yard at Kinoya (2015)



Rubbish burning on vacant land at Kinoya (2015)



Near Kinoya- this waste could be here for collection, but there is evidence of previous burning (2016)



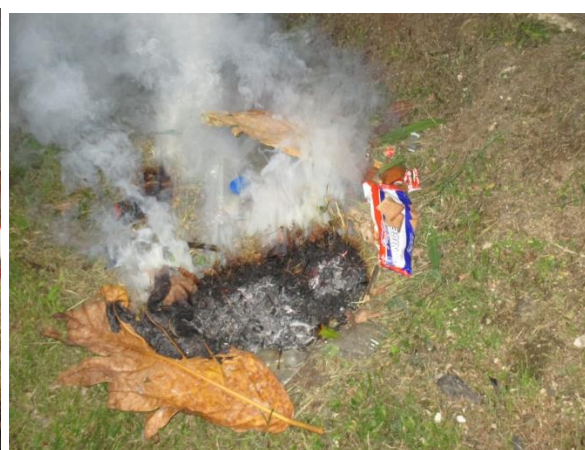
This fire, just west of Suva, smelt strongly of burning tyres (2013)



Fire is also used to clear grass and vegetation – near Suva (2013)



Caubati – burning leaves and plastic packets (2014)





Smoke from burning waste at Kinoya (2015)

Section B: Vehicle emissions



Vehicle exhaust is emitted directly onto other vehicles in Suva (2016)



This bus in Nadi shows that vehicle exhaust is also a problem there (2013)



Trucks emit smoke in Suva (2013 left and 2015 right)



Small vehicles in Suva also emit visible smoke (2015)



Drivers and pedestrians are exposed to exhaust emissions



Road and vehicle maintenance (Suva, 2016) is generally poor in Fiji, exacerbating exhaust and non-exhaust vehicle emissions (from vehicle movements, brake and tyre wear etc).

Section C: Overflowing skip bins in informal settlement areas, Suva



Despite the sign (top left) warning of a \$40 penalty for dumping waste, this skip bin near Caubati is often overflowing

Section D: Raised waste platforms



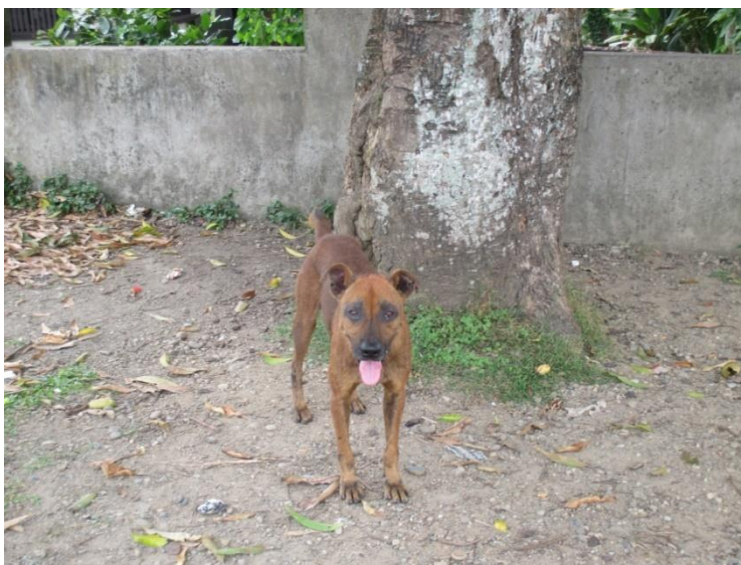
Kinoya - waste has blown off the platform onto the ground. (2016)



Lami – this platform is not high enough



Kinoya – makeshift platform (2015)
to deter dogs (2016)



Kinoya - dogs freely roam the streets in Suva, providing one reason for raised platforms and waste burning

Appendix G:

Portable samplers and flow calibration meters

1. SKC portable air sampler

Specifications from

<https://www.skcltd.com/products2/air-sampling-pumps/universal-pump.html#specifications>

The wide flow range allows dust, gas, bag and impinger sampling which when taken all together cover almost all sampling methods to meet COSHH requirements. The Universal is used by hygienists and safety professionals who demand the ultimate in flexibility and reliability.

The Universal is available as a standard "no frills" version up to a fully programmable model - all models have Intrinsic Safety and CE approval. The Universal has also found many applications in environmental and fenceline monitoring due to its timer facility.



Figure G1. SKC portable sampler

- Suitable for dust, gas/vapour, bag and impinger sampling
- ATEX certified for use in hazardous areas
- Wide flow range with built-in low flow pressure regulator

- Integral flow indicator
- High build quality for a long working life
- Clear, anti-tamper cover protects the controls
- Three models available with a choice of features

Specifications

Flow System	<ul style="list-style-type: none"> • Flow range: 5 to 4000 ml/min (5-500 ml/min requires adjustable low flow holder) • Integral Constant Pressure Controller for low flow sampling • Built-in replaceable particulate trap to prevent contamination of sample
Power Supply	<ul style="list-style-type: none"> • NiMH battery pack provides 8 hours minimum runtime at 3000 ml/min and 20 inches of water back pressure • Automatic pump shutdown in the event of a low battery condition • Low battery indication on LCD (Intermediate and Deluxe models only) • Battery eliminators available for running the pump continuously from AC mains (note that ATEX certification will be invalidated)
Certification	<ul style="list-style-type: none"> • ATEX-approved and CE marked • Intrinsic Safety EEx ia IIC T4
Operating Temperature	<ul style="list-style-type: none"> • -20 to 40°C (-4 to 104°F)
Case	<ul style="list-style-type: none"> • Metal casing with integral accessory mount and belt clip
Dimensions	<ul style="list-style-type: none"> • 13.1 x 13.1 x 5.7 cm (5.1 x 5.1 x 2.2")
Weight	<ul style="list-style-type: none"> • 915 g (32.2 oz)



Technical Note

The Use of SKC Aluminum Cyclones

In hopes of achieving worldwide consensus, new size-selective exposure guidelines for particulates have been recently adopted by various agencies including:

- International Organization for Standardization (ISO)
- Comité Européen de Normalisation (CEN)
- American Conference of Governmental Industrial Hygienists (ACGIH)
- ASTM International

One of the most significant changes was the adoption by these agencies of a 50% (or median) cut-point for a respirable particulate matter sampler of 4 μm . The National Institute for Occupational Safety and Health (NIOSH) has now also adopted this same definition in their Method 7500 for Silica, Crystalline (15 January 1998 Issue) and Method 0600 Particulates not Otherwise Regulated, Respirable (15 January 1998 issue).

In light of these developments, SKC contracted with New York University and consulted with leading aerosol research scientists to validate the SKC Aluminum Cyclone to meet the new definition of respirable particulate matter. It was determined that a flow rate of 2.5 L/min results in a collection efficiency curve that best matches the new definition.¹ See Figure G2.

Health and safety professionals in the United States, however, face a problem in that the Occupational Safety and Health Administration (OSHA) has not yet adopted the new particulate definitions. Like with the Permissible Exposure Limits (PELs), OSHA must continue to use particulate definitions and standards issued in the 1960s until updates make their way through the regulatory process.

Since OSHA compliance officers must use cyclones that meet the OSHA definition for respirable particulate matter still in effect, they cannot use the SKC Aluminum Cyclone or any other cyclone designed exclusively to meet the new definition. OSHA compliance officers are limited at this time to the use of the 10-mm nylon cyclone (Dorr-Oliver). Other health and safety professionals, however, face the choice of adopting the latest developments in science and technology or following the methods that would be used in case of an OSHA inspection.

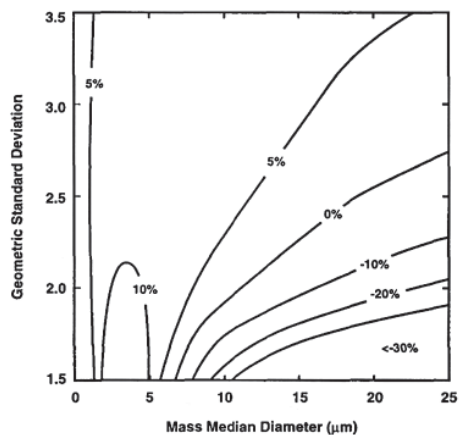


Figure G2.

Bias Map: SKC Aluminum Cyclone at 2.5 L/min¹

Users must ultimately choose the sampling equipment most appropriate for an application. However, SKC would like to provide the following information for consideration:

1. The SKC Aluminum Cyclone at a flow rate of 2.5 L/min meets the ACGIH definition of a respirable dust sampler.^{1, 6}
This is an important consideration for companies that adopt ACGIH Threshold Limit Values[®] as in-house standards.
2. The literature has reported that the sampling efficiency of the 10-mm nylon (Dorr-Oliver) cyclone is affected by the airstream velocity and inlet orientation.² Studies have shown also that the 10-mm nylon cyclone can experience as much as a 20% reduction in mass collected when sampling at an angle toward the wind.³
3. The literature suggests that the 10-mm nylon cyclone collects proportionately less dust (undersamples) with decreasing humidity than either the horizontal elutriator (as a reference instrument) or the SKC Aluminum Cyclone, as a result of electrostatic charges.⁴

OSHA silica experts feel, however, that results for crystalline silica obtained with cyclones based on the new definition may not be consistently higher than those obtained by OSHA inspectors since the OSHA PEL for compounds containing crystalline silica is based upon the percent silica content. OSHA believes that different particle classification devices collect different *distributions* of particles which would affect the percent silica content in the sample.⁵

In the years ahead, SKC hopes that all agencies involved in occupational health and safety issues will be in agreement on the definitions of particle size fractions and appropriate samplers. Until then, health and safety practitioners will need to decide how best to address this disparity in order to protect their employees from exposure to particulates.

References:

- ¹ Harper, M., C.P. Fang, D.L. Bartley, B.S. Cohen, "Calibration of the SKC Inc. Aluminum Cyclone For Operation in Accordance with ISO/CEN/ACGIH Respirable Aerosol Sampling Criteria," *J. Aerosol Sci. Vol. 29*, pp. S347-S348, 1998
- ² Kar, K. and M. Gautam, "Orientation Bias of the Isolated 10-mm Nylon Cyclone at Low Stream Velocity," *Am. Ind. Hyg. Assoc. J. 56*:1090-1098, 1995
- ³ Liden, G., "Evaluation of the SKC Personal Respirable Dust Sampling Cyclone," *Appl. Occup. Environ. Hyg. 8*(3):178-190, 1993
- ⁴ Sass-Kortsak, A.M., C.R. O'Brien, P.R. Bozek, J.T. Purdham, "Comparison of the 10-mm Nylon Cyclone, Horizontal Elutriator, and Aluminum Cyclone for Silica and Wood Dust Measurements," *Appl. Occup. Environ. Hyg. 8*(1):31-37, 1993
- ⁵ Edwards, S.L., "Crystalline Silica: Sampling and Analytical Issues," *Synergist*, Dec. 2000, pp. 11-13
- ⁶ Trakumas, S., et. al., *Performance Assessment of Personal Respirable Cyclone Samplers*, AIHce Presentation 191, 2003

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2. Ecotech Microvol

MicroVol 1100

LOW VOLUME AIR SAMPLER



The MicroVol 1100 low volume air sampler provides a flexible sampling platform for PM_{10} , $PM_{2.5}$ or TSP particulates and basic meteorological parameters.

The MicroVol 1100 is suitable for both indoor and outdoor applications. The unit is microprocessor controlled and uses a mass flow sensor in conjunction with ambient temperature and pressure sensors to automatically maintain a constant volumetric flow rate.

APPROVALS

- PM_{10} AS/NZS 3580.9.9 2006
- $PM_{2.5}$ AS/NZS 3580.9.10 2006
- Manufactured under ISO 9001.

INDOOR SAMPLING

- Low power consumption
- Quiet operation - ideal for indoor air quality studies
- Volumetric flow control automatically corrected to standard reference temperature
- Ultra-efficient, long life DC pump delivers flow rates of 1.0 to 4.5 L/min.

OUTDOOR SAMPLING

- Wind direction and speed used to activate/de-activate sampler
- Fence line monitoring available with a network of samplers
- Built for all conditions - lightweight, rugged weatherproof construction
- Can operate via battery or solar powered sources (optional).

ENHANCED COMMUNICATION

- RS232 output for data collection and remote communication
- Filter block and instrument error alarms available
- Total control of instrument remotely from PC
- Simple programming of sampling periods, including daily and weekly programs, with in built "1-in-X day" sampling capability.

DIRECTIONAL SAMPLING

- Wind direction and speed used to activate/de-activate sampler
- External trigger (0 - 5 VDC) can be used for activating sampling program.

SPECIFICATIONS

Operation:	Microprocessor controlled (internal data logging)
Volumetric flow range/accuracy:	1.0 - 4.5 L/m
Flow accuracy:	± 2 % of reading
Flow repeatability:	± 0.5 % of reading
Temperature range accuracy:	0 to 45 °C ± 1 °C
Barometric pressure range:	600 - 900 Torr ± 4 Torr
Filter types:	47 mm ringed circular filter
Inlets available:	PM ₁₀ , TSP (standard), PM _{2.5} (optional)
Sampler dimensions:	300 x 170 x 170
Sampler weight:	3.75kg
Battery pack dimensions:	185 x 170 x 170
Battery pack weight:	4.4 kg
Battery pack life:	Up to 40 hours sampling from fully charged battery pack
Operating voltage:	12 VDC
Power consumption:	2.5 - 3 watts depending on filter loading
Standard accessories:	<ul style="list-style-type: none"> • TSP/PM₁₀ size selective inlet • Single 47 mm filter holder • 100 - 240 AC to 12 VDC power converter • MicroVol Downloader software • RS232 cable

COMMUNICATION & DATA LOGGING

Number of readings

- 150 (averaging period is user selectable, e.g. 75 hrs of 30 min averages)

External inputs

- 1 x wind direction sensor input (10 k potentiometer)
- 1 x wind speed sensor input (contact closure)
- 1 x spare contact closure input (e.g. tipping bucket rain gauge)

Output

- RS232C

OPTIONS

- Purpose built battery pack, or solar panel and battery pack
- Moisture elimination system
- Optional PM_{2.5} size selective inlet adaptor
- Optional wind speed and direction sensor or tipping bucket rain gauge.



3. Gillan Gillibrator-2 primary flow calibrator



Figure G3 . Gillan Gillibrator-2 primary flow calibrator

Specifications, from

<http://www.sensidyne.com/air-sampling-equipment/calibration-equipment/gilibrator-2/>

Three bubble cells are available, for air flow ranges of 1 to 250 cc/min (low flow cell), 20 cc/min to 6 LPM (standard flow cell) and 2 to 30 LPM (high flow cell). These interchangeable wet cells generate perfect bubble films at the touch of a button. An infrared sensor reads the bubble flow rate, which is then calculated and displayed.



Figure G4: Flow cells for Gillan Gillibrator-2 primary flow calibrator

Low Flow Cell

Dimensions: 2W x 4H x 2.1"D (51W x 102H x 53mmD)

Weight: 0.4 lbs. (.18 kg)

Standard Flow Cell

Dimensions: 2.5W x 6H x 2.6"D (64W x 152H x 66mmD)

Weight: 0.82 lbs. (0.37 kg)

High Flow Cell

Dimensions: 3.5W x 8.1H x 3.7"D (89W x 206H x 94mmD)

Weight: 2.26 lbs. (1.02 kg)

Flow Range, Accuracy

Low Flow Cell, 1 - 250 cc/min, $\pm 1\%$ of reading accuracy;

Standard Flow Cell, 20 cc/min to 6 LPM, $\pm 1\%$ of reading accuracy;

High Flow Cell, 2-30 LPM, $\pm 1\%$ of reading accuracy.

Temperature Limits

Operating Temperature: 5° to 35°C (41° to 95°F);

Storage Temperature: 0° to 50°C (32° to 122°F)

Electrical

DC Power Source: Internal Battery Pack;

AC Power Source: Continuous operation through adapter/charger;

Battery Charge Time: 14 Hrs.;

Expected Battery Life: over 300 charge/recharge cycles;

Transmission Link: RS-232;

4. Shigawa DC-1 dry test gas meter

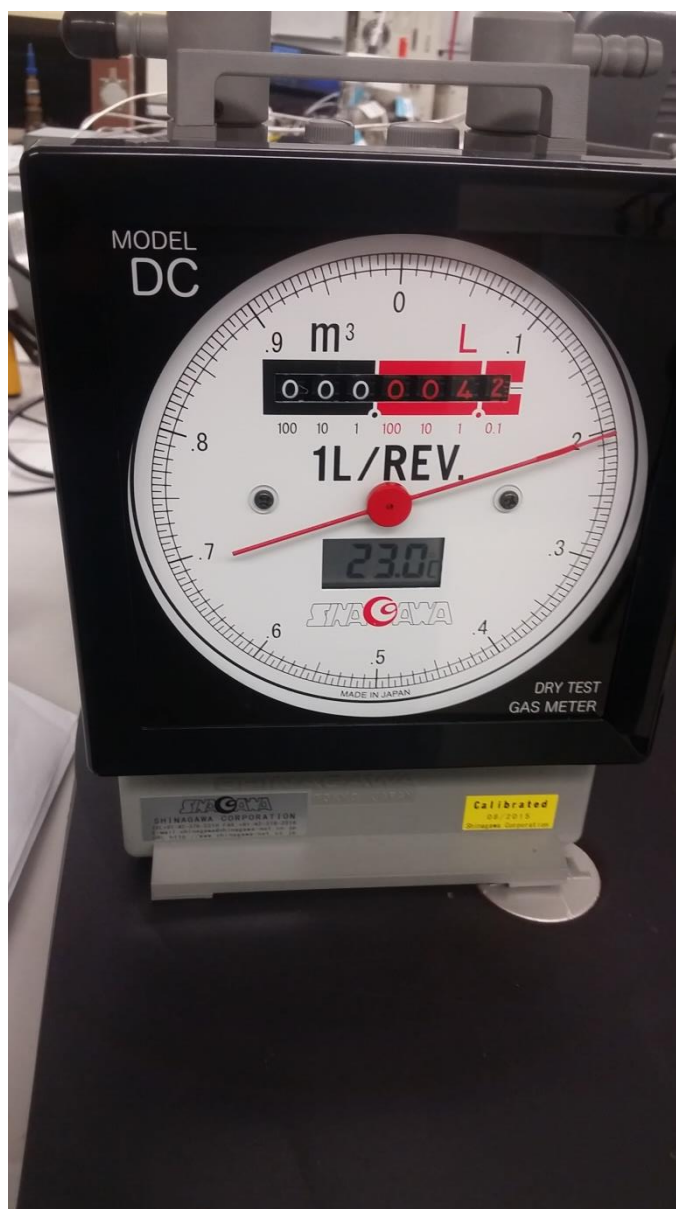


Figure G5: Shigawa DC-1 dry test gas meter

This meter is of the volumetric positive displacement type. As such, almost all types of gas can be measured without influence from relative density, viscosity, etc. It does not moisten gas, so the measurement has no influence from vapor-pressure (http://www.shinagawa-net.co.jp/e_m2_2.html).

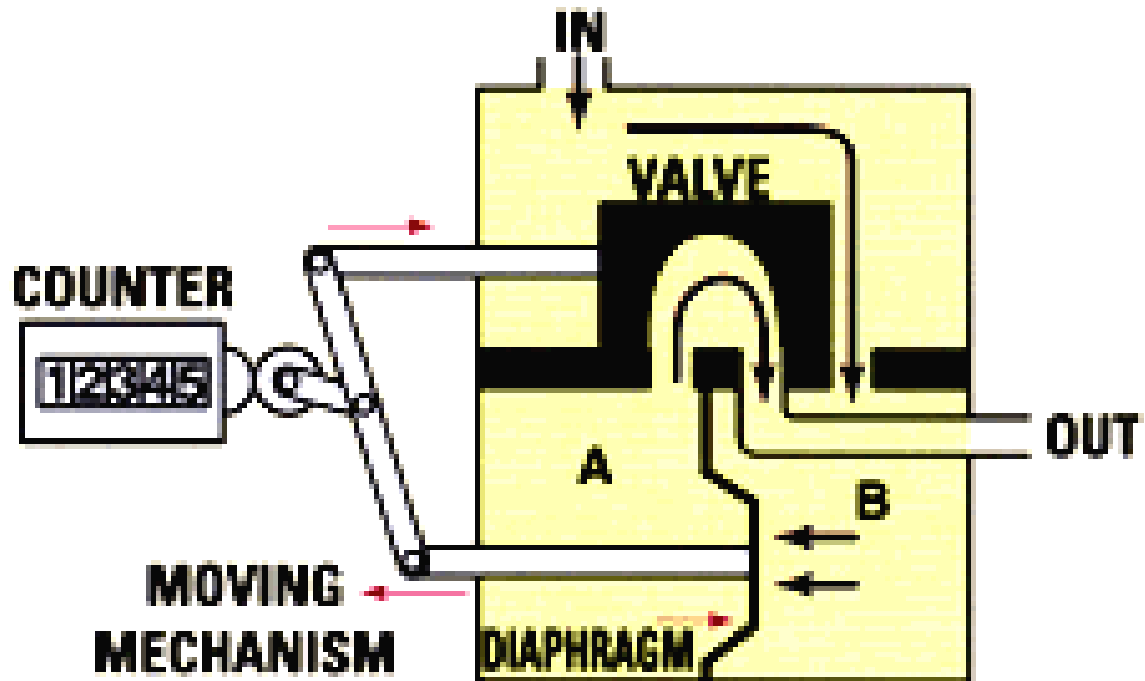


Figure G6: Function of Shigawa DC-1 dry test gas meter

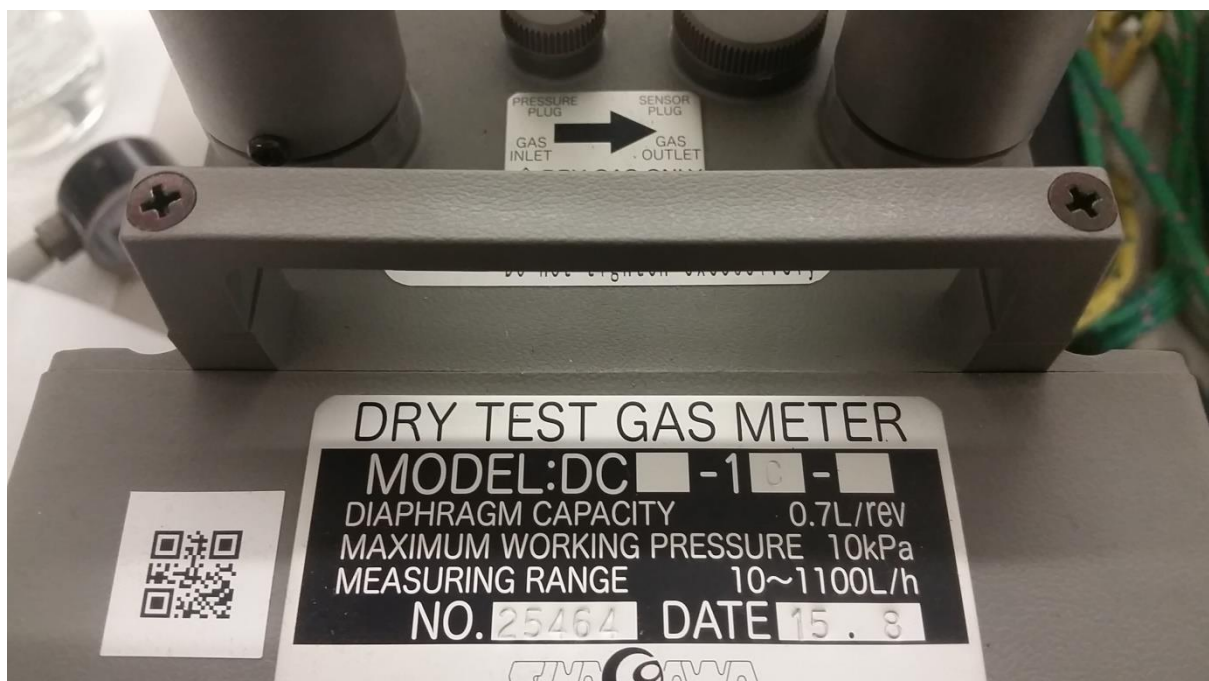


Figure G5: Specifications: Shigawa DC-1 dry test gas meter