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

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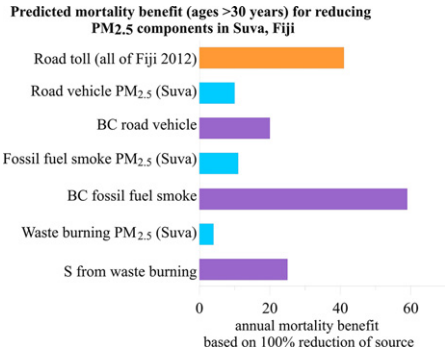
HIGHLIGHTS

- 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



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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 μg/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; Mataki, 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 ± 0.3 μg/m^3 annual average of Wednesdays and Sundays) yet black carbon in $PM_{2.5}$ (2.2 ± 0.1 μg/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°S, 178.42°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-

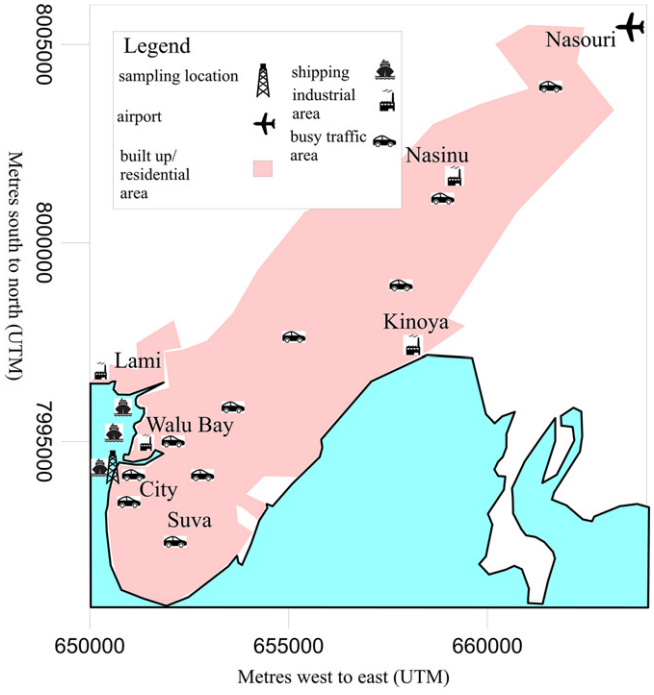

Fig. 1. Sampling locations, showing the $PM_{2.5}$ sampling location in Suva city centre as well as locations of major industries and other emission sources.

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 m^2/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 Tandetron 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 backscattering 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/m^3 (<0.02 μg/cm^2 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/m^3; ranging from 0.00025 μg/m^3 for Ni to 0.043 μg/m^3 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 $X(n, m) = factors(p,m) \times contribution(n,p) + error(n,m)$; 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

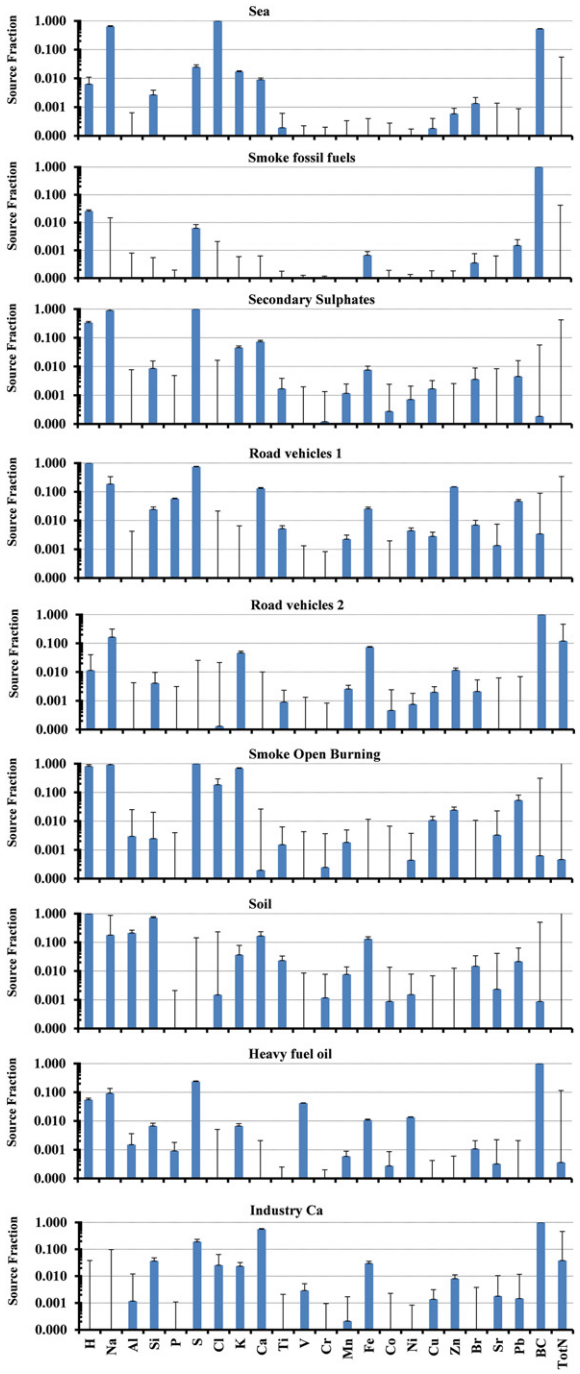

Fig. 2. Factors identified at Suva city, showing contribution of each factor to total $PM_{2.5}$ mass. The vertical axis of each graph has been normalised so that the main element in each factor has a fractional contribution value of one.

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 ± 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* (HYSPLIT) 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:

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

where:

- β mortality risk estimate as percentage increase
- Yo 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 southeast, 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.

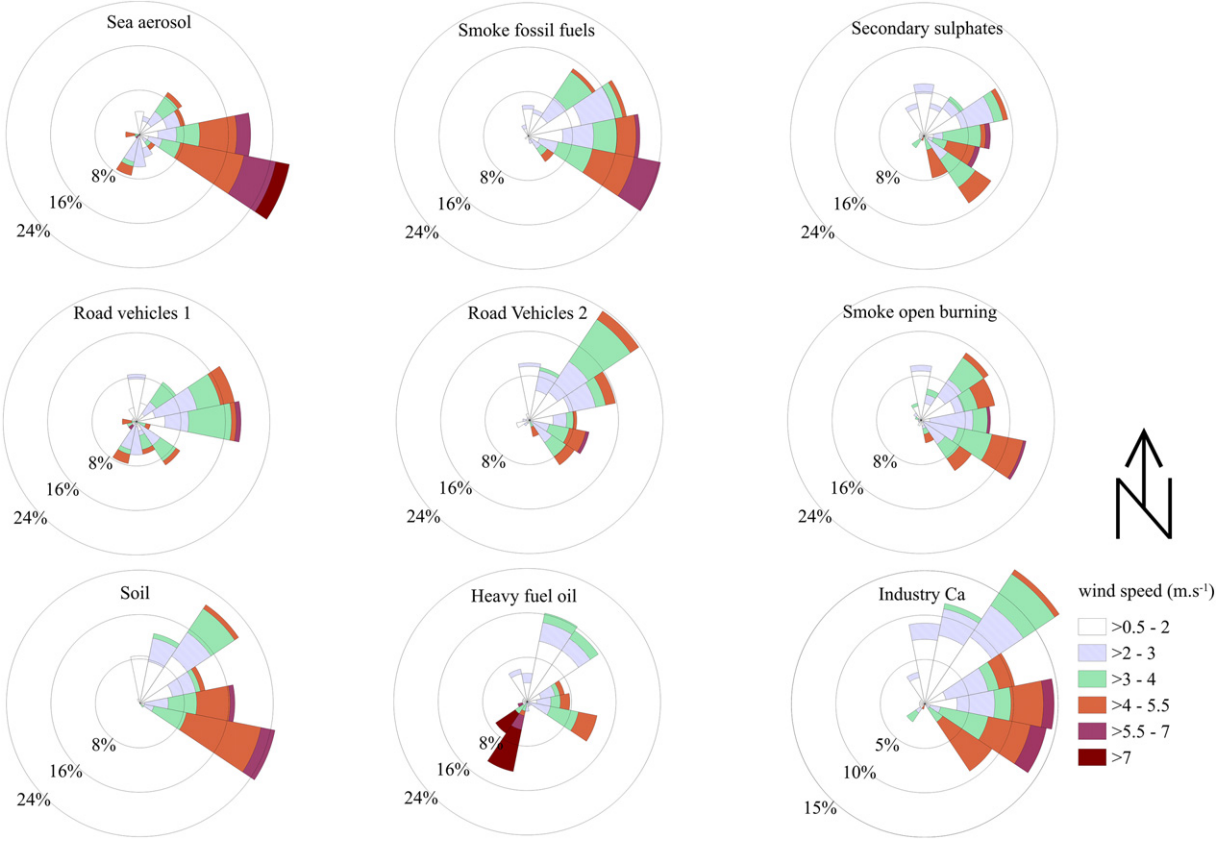

Fig. 3. Wind roses showing wind-speed and direction dependencies of source factors at Suva City. For each source, the percentage of counts for each wind direction and speed category are shown for peak 5% of occurrence of that source, based on the PMF model.

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 ± 1% of $PM_{2.5}$ 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 $PM_{2.5}$. 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ components for which health impacts have most commonly been reported, however results have varied between studies. In summary of Table 2, the risk from $PM_{2.5}$ 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 $PM_{2.5}$

Table 2
Studies 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
Roper et al. (2017)	Toxicology	Metals	Lung tissue inflammatory response results were inconsistent.
Dai et al. (2014)	Epidemiology	Ni	Ambient Ni exposure is associated with increased blood pressure
Liu (2016)	Epidemiology	As and Cu	Br, K, Na^+, and sulphate associated with emergency department visits
		Br, K, Na	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
Beelen et al. (2015)	Cohort	S	Associated with mortality (1.14; 95% CI: 1.06, 1.23 per 200 ng/m^3)
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 $\mu g/m^3$), 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
Kioumourtzoglou 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.
Kioumourtzoglou 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	Associated with cardiovascular hospitalisations, people >65 years
		Road dust, sea salt, Al, Ca, Cl, BC, Ni, Si, Ti, and V	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
			species studied to give significant association
Grahame et al. (2014)	Review	BC mainly from diesel and traffic	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% ± 3%/0.26 μg/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 NO_3^- 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 μg/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 μg/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 μg/m^3) increase; consistent with Hoek et al. (2013). Measurements of BC and EC have been shown to be approximately comparable (BC:EC = 1 ± 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 μg/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 μg/m^3 EC increase); a rate of 1.021 (1.00, 1.06) per 0.26 μg/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 μg/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 μg/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 μg/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 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.

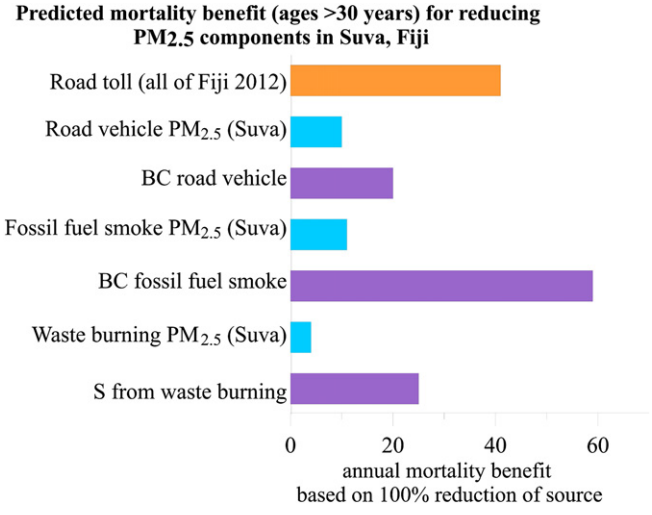

Fig. 4. Comparison of potential annual mortality reduction from reduction of different $PM_{2.5}$ sources in Suva Fiji. These potential benefits are shown in terms of risk based on $PM_{2.5}$ mass alone and risk based on specific $PM_{2.5}$ components. These are compared to Fiji's road toll in order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

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 $\mu g/m^3$ 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

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.08.225.

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