Source Apportionment of Ultrafine and Fine Particle Concentrations in Brisbane, Australia

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Abstract

Purpose: To investigate the significance of sources around measurement sites, assist the development of control strategies for the important sources and mitigate the adverse effects of air pollution due to particle size.

Methods: In this study, sampling was conducted at two sites located in urban/industrial and residential areas situated at roadsides along the Brisbane Urban Corridor. Ultrafine and fine particle measurements obtained at the two sites in June-July 2002 were analysed by Positive Matrix Factorization (PMF).

Results: Six sources were present, including local traffic, two traffic sources, biomass burning, and two currently unidentified sources. Secondary particles had a significant impact at Site 1, while nitrates, peak traffic hours and main roads located close to the source also affected the results for both sites.

Conclusions: This significant traffic corridor exemplifies the type of sources present in heavily trafficked locations and future attempts to control pollution in this type of environment could focus on the sources that were identified.

Keywords: *Positive Matrix Factorization, Particle number concentration, Urban corridor, Source identification*

1. Introduction

The health effects of airborne particulate matter have been studied extensively and reviewed in recent papers for inorganic and organic chemical composition as

well as for the effect of the size of the particles (Pope et al. 2002; Mauderly and Chow 2008; Schlesinger 2007). Lippmann et al. reviewed studies of the impact of ambient particulate matter and determined that there needs to be further studies involving continuous monitoring of pollutants to determine their short term health effects (Lippmann and Chen 2009; Lippmann 2009). There is still a debate on whether the chemical composition, size or the surface area of particles is the most relevant predictor of their health effects (Kasumba et al. 2009; Lighty et al. 2000). Therefore, it is important that the identities, locations, and impacts of the sources for the different types of information are determined. As well as finding the links between a source and the health impacts, air quality control strategies can be developed.

Airborne particulate matter (PM) is routinely monitored in many countries and this has led to standards for PM with less than 10 µm and 2.5 µm aerodynamic diameters (PM_{10} and $PM_{2.5}$, respectively). In Australia, to regulate the concentrations of pollutants, standards for air quality were created under the National Environmental Protection Measures (NEPM) in 1998 and updated in 2003 to set targets for concentrations of pollutants such as gaseous species (CO, NO_2, O_3, SO_2), elements (Pb) and particle mass (PM_{10}, $PM_{2.5}$) but not for particle size (NEPC 1998, 2003). Particulate matter can be separated into coarse, fine, ultrafine and nanoparticles based on the size. Nanoparticles (<50 nm, arbitrary definition, not internationally accepted definition) (Morawska et al. 2008) are often formed after emission of semi-volatile particles as precursors to secondary particles. Ultrafine particles (<100 nm, arbitrary definition) are emitted mostly from motor vehicles (Ogulei et al. 2007b), while fine particles (accumulation mode 100 nm -1.0 μm) are formed by chemical reaction. Fine and ultrafine particles are of concern due to the health effects that may occur when they deposit in the lungs. Finally, coarse particles (1-10 μm) are aerosolized mechanically by the abrasion of road materials (Wahlin et al. 2001) sea spray productions, and also biological materials, including pollens, leaves and insects. Because of the different processes occurring in the atmosphere, these classes of particles change dynamically (Kim et al. 2004). Zhu et al. (2002) showed that the particle sizes change as the distance to a road varies (Zhu et al. 2002). However, the

distributions can be assumed to be stable because the time, distance and changes from source to receptor site are consistent..

Particles measured at a receptor site are mixtures of emissions from different sources. Therefore, by applying chemometric techniques, the relevant sources of the particle numbers can be determined. Being a source apportionment model, Positive Matrix Factorization (PMF) can be used to resolve the dominant factors from measurements made at a receptor site. Thus the model has previously been used extensively for the source apportionment of ambient particulate mass (Reff et al. 2007; Lee et al. 1999; Song et al. 2001; Begum et al. 2004; Kim et al. 2003b; Friend et al. 2011a; Friend et al. 2011b). PMF has the advantages of weighting the data points individually and applying a non-negativity constraint to the outcomes of the analysis. Previous application of the model was largely for particle composition data. For particle size distribution data, PMF was first applied in Seattle, Washington (Kim et al. 2004), where PMF and another receptor model, UNMIX, were applied to 3 months of measurements. That paper correlated the results with some chemical species and also indicated that particle size emissions are stable enough for factor analysis. Similar analysis has also been conducted in Erfurt, Germany (Yue et al. 2008). In addition, there have been other types of analyses including, the use of (i) gaseous, chemical (particulate matter composition), and particle size data together in the PMF analysis over short (Ogulei et al. 2006b; Zhou et al. 2005a; Thimmaiah et al. 2009), or long (Ogulei et al. 2007a; Kasumba et al. 2009; Cuccia et al. 2010) periods, (ii) chemical and particle size data in two separate PMF analyses followed by comparison of the outcomes (Gu et al. 2011), (iii) elemental size distribution data (Han et al. 2006; Mazzei et al. 2007), (iv) aerodyne aerosol mass spectral data (Sun et al. 2010), and (v) PMF results that are compared to the actual events observed during the sampling period (Ogulei et al. 2007b). Numerous sources have been identified in these studies such as: nucleation, various motor vehicle sources, wood burning, and secondary particles.

In Brisbane, extensive studies have been performed on particle number size distributions in both indoor and outdoor air. Vehicle emissions have been a significant focus of the work which included a review of the characteristics of

exhaust emissions (Morawska et al. 2008). Other sources that have been investigated included biomass burning and industrial emissions (Ristovski et al. 2010; Morawska et al. 2006). Although these studies have provided information on various types of emissions, source apportionment was not specifically performed on any of the resulting data. However, PMF analysis has been performed on chemical compositional data obtained in this area (Friend and Ayoko 2009).

The objective of the current study was to apply Positive Matrix Factorization to particle number size distribution data from two roadside sites located in urban/industrial and residential areas, respectively, in Brisbane Australia. Results of these analyses reveal possible sources of the particles that can be compared with previous data collected at the sources. Thus the contributions of the sources to the detected size distributions were estimated and the time variations of the source emissions were then examined.

2. Experimental Methods

2.1 Sample Collection

The data used in this study were collected from June-July 2002 in some of the southern suburbs of Brisbane. This area is part of the Brisbane Urban Corridor defined by the Australian government as containing major links between the areas and the city, and includes the major highways and motorways in the area. One of the identified roads links the Ipswich Motorway and the Gateway Motorway and is named at different points along the road as Granard Road, Riawena Road, Kessels Road and Mt Gravatt–Capalaba Road (Figure 1). Along with the previously mentioned motorways, Beaudesert Road and the Pacific Motorway also cross the link road. The chemical concentration and meteorological data were measured only over a shorter sampling period compared to the particle number size distribution data. The two sampling sites had the following descriptions.

Data from the first site was collected on Mount Gravatt-Capalaba Road between Newham Road and Logan Road. Situated at Latitude -27.5559º and longitude 153.0902º, this site was in a residential environment at the bottom of a small

valley with stop-start traffic on nearby roads and was upwind from the main roads in the area. The site was located 3 m from the road. PM_{10}, CO, NO, NO_2, wind speed and wind direction, were measured between 10:55 AM on 13 June 2002 and 11:40 PM on 14 June 2002 for the first site (433 samples).

The second site was close to a major sports stadium. This site was an urban/industrial site located at Latitude -27.5607º, Longitude 153.0658º on Kessels Road at Mains Road with semi-open terrain and it is located 2 m from the road. The stadium has a capacity of 48,500 and was home to a local football team which played a match with ~ 20,000 in attendance during the sampling period. Biomass burning was performed in the region during the period. PM_{10}, CO, NO, NO_2, wind speed and direction, were measured between 11:00 AM on 2 July 2002 and 7:00 AM on 3 July 2002 (241 samples) at the site.

2.2 Measurement Instrumentation

Scanning Mobility Particle Sizers (SMPS TSI Model 3071) were used for the determination of particle number size distributions. The detectable particle size at the site ranged from 0.014 to 0.715 µm including 110 size bins with a complete size scan every 5 minutes, while the second site had 109 size bins. Tapered Element Oscillating Microbalance (TEOM) units were used for PM_{10}. Measurements of CO, NO and NO_2 concentrations were also made. The instruments were located in a mobile trailer placed 2 to 3 m from the roadside with the sample intake 2 to 3 m above the ground. Meteorological parameters, wind speed and direction, were only measured at Site 1. The wind direction was almost exclusively from the north-west or south-west. Site 1 sampling took place between 4:10 PM on 6 June 2002and 11:42 PM on14 June 2002 resulting in 2040 samples after removal of some data points due to extreme values. However, 1629 samples were collected between 11:00 AM on2 July 2002 and 9:25 AM on 8 July 2002 at Site 2. To determine the sources of the particles detected at the site, advanced chemometric techniques were applied.

2.3 Positive Matrix Factorization (PMF)

PMF was developed by Paatero (Paatero and Tapper 1994; Paatero 1997) due to problems inherent to eigenvector-based methods. In this analysis, the PMF2 program was used and run in the robust mode, which decreases the weights of outlying data points, and allows a robust factorization to be obtained (Paatero 1997). Receptor models determine the factors that are responsible for the data measured at the sampling sites. The model is based on the mass balance equation that requires that the concentrations detected at the sampling site are consistent with the source. Kim et al. 2004 suggested that the sources of particles produce characteristic distributions and that the emissions are stable enough in the atmosphere to permit factor analysis to be performed.

An advantage of the model is the use of uncertainties for individual data points. In this study, uncertainties were calculated using the following equation (Ogulei et al. 2007a):

$$\sigma_{ij} = 0.01\left(x_{ij} + \bar{x}_j\right) \quad (1)$$

where σ_{ij} is the uncertainty estimated for size bin j and sample i, 0.01 is an arbitrary constant empirically determined by Ogulei through trial-and-error, x_{ij} is the observed number concentration, and $\bar{x}_j$ is the arithmetic mean of the values from size bin j. The overall data point uncertainty was calculated as

$$s_{ij} = \sigma_{ij} + C_3 \max\left(\left|x_{ij}\right|, \left|y_{ij}\right|\right) \quad (2)$$

where y_{ij} is the calculated value for x_{ij}, and C_3 is a constant.(Ogulei et al. 2006a) The rotational parameter FPEAK is used to impose rotations on the solutions in order to obtain more physically reasonable results (Moon et al. 2008).

Another characteristic of the model is that the data entered must be non-negative. So, missing values were replaced with the geometric mean of the size bin and assigned an uncertainty that is four times the mean.

To determine the ideal number of factors for the analysis, the Q (theoretical number of points in the data matrix) was used and the standardised residuals were examined to ensure that the values for each size fraction were as much as possible normally distributed between -3 and 3 (Buzcu-Guven et al. 2007). The most significant aspect in determining the number of factors is how physically reasonable the identified factors are by comparing the results with previous studies that obtained samples directly from the sources, or applied PMF to similar data in order to identify the sources of the pollutants.

The results obtained are scaled to the measured particle numbers by regression of the total number concentration against the obtained source contributions to determine a scaling constant, s_k (Ogulei et al. 2007a; Kim et al. 2003a).

$$x_{ij} = \sum_{k=1}^{p} (s_k g_{ik})(f_{kj} / s_k) \qquad (3)$$

2.4 Data treatment

Two separate PMF analyses were carried out, one with the data matrix with the full particle number size distributions and the other with only the distribution data that was collected at the same time as the chemical and meteorological data. Correlations were then performed for the g_{ik} and f_{kj} matrices between the two PMF runs to ensure that the sources identified were the same.

After the PMF analyses had been completed, the contributions of the sources found during the partial particle distribution PMF analyses were compared with the measured concentrations of the chemical species using Pearson correlation coefficients (Kim et al. 2004; Gu et al. 2011).

2.5 Conditional Probability Function (CPF)

CPF analysis is performed in order to determine the likely locations of the sources. By combining the contribution results from the PMF analysis with the meteorological measurements (wind speed and direction), the directions of the sources from the sampling site were ascertained (Lee and Hopke 2006). CPF is defined by:

$$\text{CPF} = \frac{m_{\Delta\theta}}{n_{\Delta\theta}} \quad (4)$$

Where $m_{\Delta\theta}$ is the number of events from wind sector $\Delta\theta$ which are greater than the 75th percentile of the fractional contribution from each source, and $n_{\Delta\theta}$ is the total number of events from the same wind sector (Kim and Hopke 2004). Wind direction bins ($\Delta\theta$) were set at an angle of 24 degrees in this study so that there were 15 wind sectors. If insufficient samples (less than 10) were contained within a wind direction bin $n_{\Delta\theta}$, the values were discarded (Yue et al. 2008). Calm wind speed samples (<1 m/s) were excluded from the analysis. The sources are likely to be located in the direction with high CPF values.

3. Results and discussion

The gaseous species measured at the sites were compared with the Australian standards (NEPC, 2003) and it was determined that the concentrations of CO and NO_2were below their respective Australian Standards. Summary Statistics for the chemical and particle size data are shown in Online Resource 1 and 2.

Six factors were identified as the best number of sources at both Site 1 and Site 2. These results were based on several parameters. The Q values obtained were compared with the theoretical values. The FPEAK values were varied from -0.5 to 0.5 and evaluated using the G space plotting approach to explore the rotational space and identify the edges of the data. An FPEAK of 0 was found to correspond to good G space plots for both sites (Ogulei et al. 2006a). The two PMF analyses that were performed for the full particle number concentration data and for the

partial concentrations that corresponded with the other data were compared by correlating the G and F factors. Significantly stronger correlation values were obtained between the factors that were identified as the same than for any other factor. Finally, the results were compared with previous studies in the literature that measured the sources directly or performed PMF analysis on similar data.

The results for the source profiles at both sites are displayed in Figures 2 and 3. Contributions from the PMF analyses are displayed in Online Resource 3 and 4 while the diurnal variations (the average values for each hour of the day) are presented in Online Resource 5 and 6. For Site 2, the contributions showed a similar pattern for all of the sources, so, the chemical species correlations also showed similar trends for all of the sources. Graphs of the correlations between the PMF contributions and the measured chemical species concentrations are provided in Online Resource 7 and 8. Online Resource 9 shows the CPF results with an example of the standardised residuals for Site 2 provided in Online Resource 10. The source profile plots for the reduced PMF analysis with particle size samples that corresponded with when the gaseous, chemical, and meteorological data was collected are provided as Online Resource 11 for site 1 and Online Resource 12 for site 2. Table 1 shows a summary of the graphs provided in the Online Resource. The percentage contribution values are presented in Table 1 and represent the significance of a source compared to the total contributions found for all of the sources combined.

Five of the six sources were identified to be common to both sites. The source profile results were very similar for the sources, but there were some differences for the other graphs. The correlations with chemical species graphs showed that for site 1, PM_{10} was significant for almost all of the sources, whereas for site 2, NO and CO had the highest correlation for all of the sources. This may be due to the similar contribution plots affecting the chemical correlations.

The first factor showed a peak in the particle number size distribution at 20 nm. Previous papers have identified similar factors as local traffic (Gu et al. 2011; Zhou et al. 2004; Zhou et al. 2005b). While the Site 1 factor peaked at 20 nm agreeing with the literature, a source that peaked below the smallest measured

size fraction of 15 nm was found at Site 2. Site 2 was located near a forest that had significant biogenic volatile organic compound emissions that may have formed a mode in the nucleation region. There was a very strong correlation between the sources identified as local traffic at both sites. PM_{10} and NO_2 showed the highest correlation with the contributions at Site 1, while for Site 2, correlations with NO and CO were the highest. The average contributions peak observed between 6 – 9 AM (1510 cm^{-3} at Site 1 and 31845 cm^{-3} at site 2) agrees with the assignment of the source as local traffic. With a 30% contribution for Site 2, this factor is the highest contributing source, but it is only 15% and the third highest contributor at Site 1. Finally, the CPF shows the direction of the source to be the south-west of Site 1 where the south-east freeway, Logan road, and Newnham road all converge together, again indicating local traffic as the source.

The peak concentrations in the second factor were between 50 and 70 nm at Site 1 with the peak occurring at slightly smaller sizes at Site 2 (50 – 60 nm). This source was called Traffic 1 and attributed to diesel traffic, including distant traffic when compared to literature analyses (Ogulei et al. 2007a). Correlation was observed for NO, CO and PM_{10}. A morning peak of 2240 cm^{-3} (6 – 8 AM) and evening of 2512 cm^{-3} (5 PM) diurnal pattern was observed for Site 1 consistent with a traffic source. While an extended period between 6 – 11 AM (14817 cm^{-3}) was observed at site 2.This source explains 28% of the total contributions at Site 1 while at Site 2, only 14% was explained. The CPF analysis indicated the direction of the source to be south-west of Site 1, again toward the significant roads in the area.

The third source had a peak in the particle number size distribution at 30 nm and was very consistent for both of the sites. An assignment of Traffic 2 was given for this source while emissions from petrol vehicles have previously been identified as having a peak at this size range (Ogulei et al. 2007a; Kasumba et al. 2009). PM_{10} and NO_2 had weak correlations with the contributions at site 1, while NO and CO correlated at site 2. The diurnal variation showed peaks of 3258 cm^{-3} at 7 – 8 AM and 2313 cm^{-3} at 5 PM for site 1 and 26414 cm^{-3} at 6 – 9 AM for site 2, again consistent with traffic emissions. This source was the highest contributor to

the overall results. CPF analysis identified the main roads to the west- south-west as the source of the emissions similar to the previous two sources.

Biomass burning or wood burning was identified as the fourth source with a peak in the particle number size distribution at 100-200 nm (Kasumba et al. 2009; Ogulei et al. 2007a). Again the result for the Site 2 analysis showed a much wider peak than that for Site 1. This may be due to significant forest back-burning that was performed on a nearby mountain during the sampling period. NO, CO and PM_{10} were found to have strong correlations for site 1 with NO and CO for site 2. The diurnal pattern again showed a peak from 6 – 8 AM but with significantly increasing values after 5 PM in the evening for site 1 and from 6 – 11 AM at site 2. CPF analysis showed a more westerly direction of the source with a peak to the north. Approximately 20% of the total contribution was attributed to this source for Site 1 and 11% for Site 2.

A bimodal distribution with peaks at 50–60 nm and 200–300 nm was found in factor 5 but was difficult to identify and so source five was called Unknown 1. A peak at 200 nm has been attributed to cloud processing with marine aerosol in Brisbane (Thomas and Morawska 2002) but there were no noticeable fog days during the sampling. Another possible source with similar distributions is secondary aerosol (Gu et al. 2011; Ogulei et al. 2007a; Kasumba et al. 2009; Yue et al. 2008). However, the sampling took place during winter when there would be less photochemical activity. Kasumba et al. 2009 showed a very similar source that was identified as secondary nitrate. This may be more reasonable due to the gas-to-particle equilibrium that could indicate higher concentration of nitrate in winter. Gu et al. 2011 identified a source that had a peak at 300 nm as re-suspended dust but a peak with > 300 nm diameter was measured in this study and no nanoparticle peaks were observed. A traffic related source was found by Kim et al. 2004, with peaks at 40-50 nm and 300 nm. The source profile results for Site 2 shows similar patterns but with a much wider peak than that for Site 1; this may be due to the occurrence of significant biomass burning during the sampling which provided larger particle sizes and influenced this source.

The highest correlations for Factor 5 were with NO and PM_{10} at site 1 and NO and CO at site 2. Again the highest peak of 443 cm^{-3} in the diurnal analysis was between 6 and 9 AM for site 1 and a value of 11481 cm^{-3} was determined for site 2. This source was found to have a significantly higher percentage contribution (10%) for Site 2 than for Site 1 (4%). Previously, it was noted that Site 2 seemed to be affected by secondary particles during the day and this may agree with that assessment. Peaks to the south and west of the sampling site were identified from the CPF analysis. These peaks are similar to those observed in previously described factors because they point to the nearby road. However, there was a peak to the northeast that may be consistent with marine aerosol.

The final source was different between the sites possibly indicating a local influence. At Site 2 there was only a single peak at greater than the maximum size bin of 700 nm. The analysis of samples collected at Site 1 showed an additional peak at 40 nm. This source was similarly difficult to identify as the preceding factor and so was called Unknown 2. As with the previous source, marine aerosols may have an impact in the higher size ranges. In Brisbane, marine aerosols are influenced by on-shore winds that carry the salt over long distances (Thomas and Morawska 2002). Comparisons of the contributions and source profiles showed small correlation between the two sites indicating local sources may be responsible. Particle sizes around 700 nm are in the accumulation mode while the peak at 40 nm is in the same size range as the motor vehicle sources. There were no significant fog events during the sampling that might explain the peak observed at higher size ranges. NO, and CO were weakly correlated with this source while the diurnal pattern was significantly different from any other source. From 1 PM to midnight, the average was consistent and higher than the morning with a peak concentration of 208 cm^{-3}. At site 2 however, similar results to the other sources were observed with a diurnal pattern peak at 6 – 10 AM (9283 cm^{-3}) and a chemical correlation with NO and CO. The CPF was also different with a strong peak to the south with a north-east peak also present possibly indicative of a marine aerosol influence. With 2% contribution at Site 1 and 9% contribution at Site 2, this source was the lowest contributor at both sites. Site 1 was located further from an intersection and this may explain the differences in the percentage contribution and in the profiles. The 40 nm peak in

the profile for Site 1 may indicate that this source is combined with the vehicle emissions because of the greater distance to the nearest intersection.

Since the two sites were separated by 2.5 km in distance and the measurements were conducted at different times, an attempt was made to analyse a data matrix which contained the particle size distributions for both sites. The source profile results obtained were very similar to those found for Site 1 but the contribution results were affected by the differences between the sites. While Site 1 was upwind of the intersection, Site 2 was downwind. Therefore the results from the separate analyses appeared to be more representative of the sites.

4. Summary

Particle number size distribution, chemical species, and meteorological data were collected at two sites in the Brisbane Urban Corridor. By applying Positive Matrix Factorization (PMF), and Conditional Probability Function (CPF) to the particle number size distribution and meteorological data, the sources of the particles were determined. Examination of the trends of the results revealed more information about the sites. Six sources were found at both sites and identified as local traffic, two traffic sources, biomass burning and two unknown sources. Gaseous nitrogen species correlated with most of the sources and peak traffic hour trends were identified in the results. The main roads close to the sites were attributed as the location of most of the sources and the traffic factors showed the highest contributions. These results are indicative of prominent characteristic emissions generated along a significant traffic corridor.

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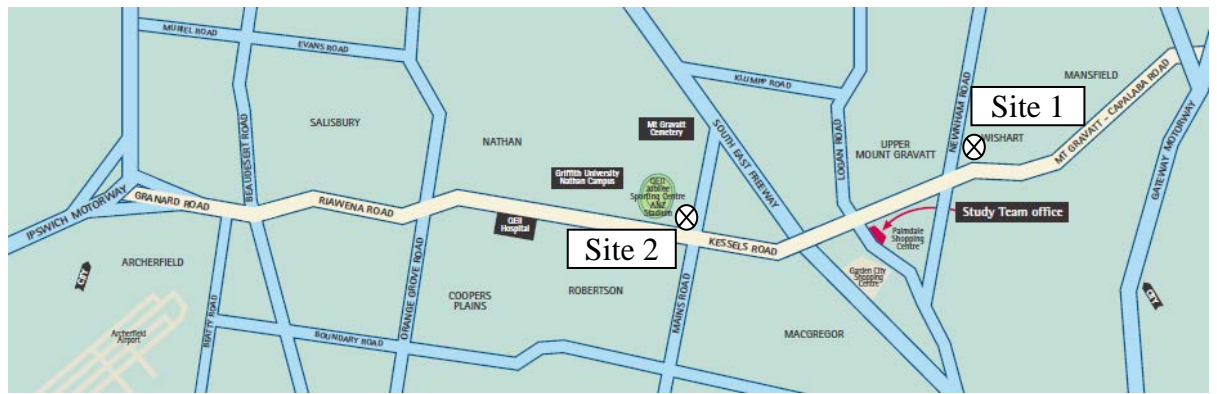
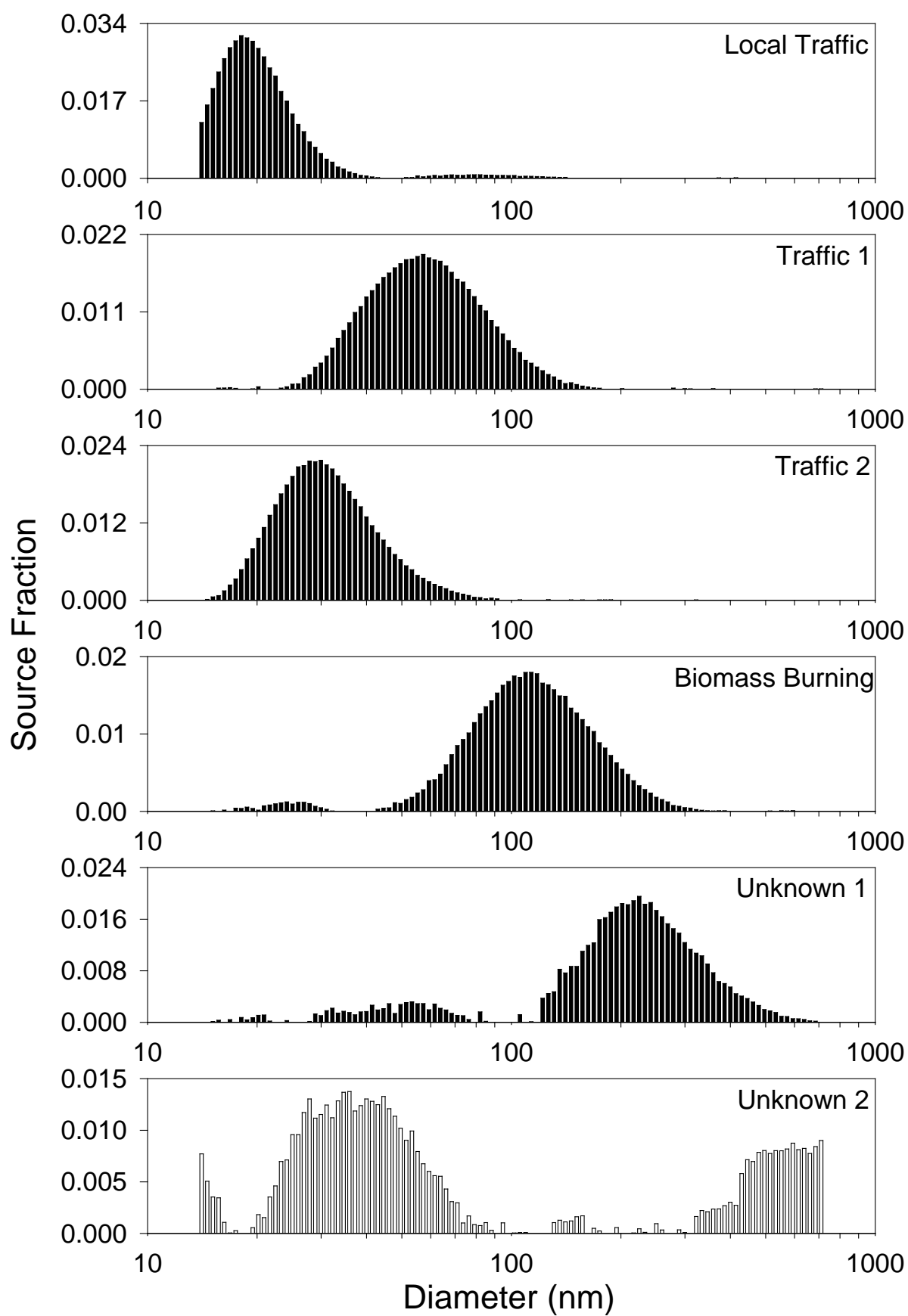Figure 1: Map of the Brisbane Urban Corridor (BUC) indicating the location of the sampling sites.

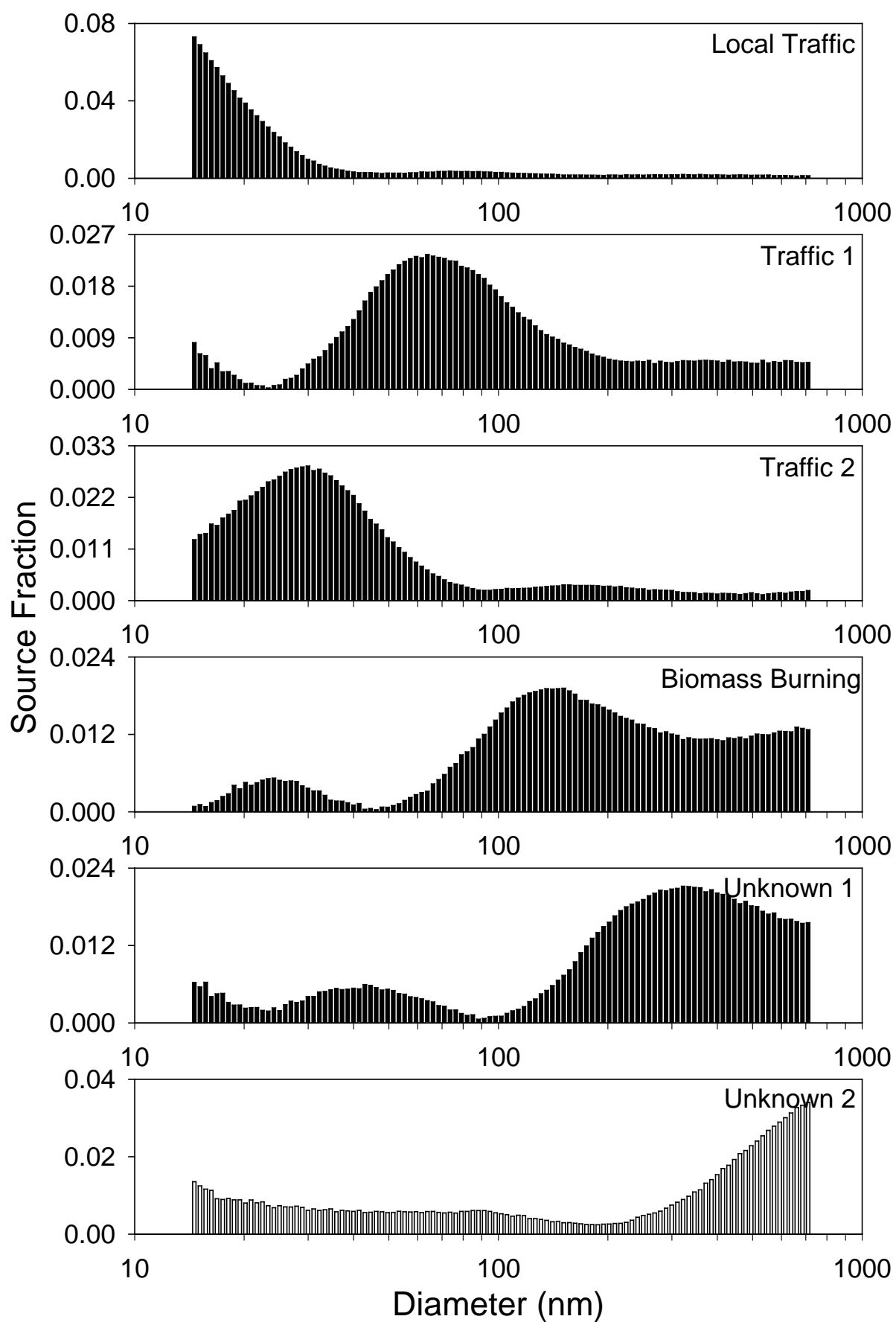Figure 2: Source Profile from the PMF analysis of Site 1.

Figure 3: Source profile from the PMF analysis of Site 2.

Source	Diurnal Variation (Peak Concentration)		Chemical Species Correlation		CPF Direction	Percentage Contribution	
	Site 1	Site 2	Site 1	Site 2		Site 1	Site 2
Local Traffic	6 – 9 AM (1510 cm^{-3})	6 – 9 AM (31845 cm^{-3})	PM$_{10}$ and NO$_2$	NO and CO	South–West	14.9%	29.7%
Traffic 1	6 – 8 AM (2240 cm^{-3}) and 5 PM (2512 cm^{-3})	6 – 11 AM (14817 cm^{-3})	PM$_{10}$ and NO	NO and CO	South–West	28.1%	14.2%
Traffic 2	7 – 8 AM (3258 cm^{-3}) and 5 PM (2313 cm^{-3})	6 – 9 AM (26414 cm^{-3})	PM$_{10}$ and NO$_2$	NO and CO	West and South–West	30.8%	26.2%
Biomass Burning	6 – 8 AM (1703 cm^{-3})	6 – 11 AM (11481 cm^{-3})	NO, CO, and PM$_{10}$	NO and CO	West and North	20.1%	11.0%
Unknown 1	6 – 9 AM (443 cm^{-3})	7 – 10 AM (11663 cm^{-3})	PM$_{10}$ and NO	NO and CO	South and West	3.9%	10.4%
Unknown 2	1 – 11 PM (208 cm^{-3})	6 – 10 AM (9283 cm^{-3})	NO and CO	NO and CO	South and North–East	2.1%	8.5%

Table 1: Summary of Results. Diurnal trends with the peak concentration in brackets, chemical species with the highest Pearson Correlation value between the species and the source contributions, the percentage contributions for the identified sources, and the direction of the source as indicated by the CPF analysis performed for site 1.