Atmospheric Research 194 (2017) 178–189

Contents lists available at ScienceDirect

Atmospheric Research

journal homepage: www.elsevier.com/locate/atmosres

Estimate of main local sources to ambient ultrafine particle number concentrations in an urban area

Md Mahmudur Rahman[a], Mandana Mazaheri[a], Sam Clifford[a,b], Lidia Morawska[a,*]

[a] International Laboratory for Air Quality and Health, Institute of Health and Biomedical Innovation, Queensland University of Technology, GPO Box 2434, Brisbane, QLD 4001, Australia

[b] ARC Centre of Excellence for Mathematical and Statistical Frontiers, Queensland University of Technology, GPO Box 2434, Brisbane, QLD 4001, Australia

ARTICLE INFO

Keywords:
Ambient ultrafine particles
Bayesian statistical model
Non-traffic sources
Nucleated particle formation
Urban area

ABSTRACT

Quantifying and apportioning the contribution of a range of sources to ultrafine particles (UFPs, D < 100 nm) is a challenge due to the complex nature of the urban environments. Although vehicular emissions have long been considered one of the major sources of ultrafine particles in urban areas, the contribution of other major urban sources is not yet fully understood. This paper aims to determine and quantify the contribution of local ground traffic, nucleated particle (NP) formation and distant non-traffic (e.g. airport, oil refineries, and seaport) sources to the total ambient particle number concentration (PNC) in a busy, inner-city area in Brisbane, Australia using Bayesian statistical modelling and other exploratory tools. The Bayesian model was trained on the PNC data on days where NP formations were known to have not occurred, hourly traffic counts, solar radiation data, and smooth daily trend. The model was applied to apportion and quantify the contribution of NP formations and local traffic and non-traffic sources to UFPs. The data analysis incorporated long-term measured time-series of total PNC ($D \geq 6$ nm), particle number size distributions (PSD, $D = 8$ to 400 nm), $PM_{2.5}$, PM_{10}, NO_x, CO, meteorological parameters and traffic counts at a stationary monitoring site. The developed Bayesian model showed reliable predictive performances in quantifying the contribution of NP formation events to UFPs (up to 4×10^4 particles cm^{-3}), with a significant day to day variability. The model identified potential NP formation and no-formations days based on PNC data and quantified the sources contribution to UFPs. Exploratory statistical analyses show that total mean PNC during the middle of the day was up to 32% higher than during peak morning and evening traffic periods, which were associated with NP formation events. The majority of UFPs measured during the peak traffic and NP formation periods were between 30–100 nm and smaller than 30 nm, respectively. To date, this is the first application of Bayesian model to apportion different sources contribution to UFPs, and therefore the importance of this study is not only in its modelling outcomes but in demonstrating the applicability and advantages of this statistical approach to air pollution studies.

1. Introduction

While ultrafine particles (UFPs, < 100 nm) contribute little to the total mass concentration of airborne particles in urban areas their number concentration is very high (Morawska et al., 2008). The role of UFPs in urban air quality, as well as human exposure to them and their subsequent health impacts, have recently been of great interest (Buonanno et al., 2011a; Buonanno et al., 2011b; Buonanno et al., 2012a; Buonanno et al., 2012b; HEI, 2013; Lim et al., 2012; Mazaheri et al., 2013b; WHO, 2013).

In general, key sources of ambient particles in urban areas are vehicular traffic, local non-vehicular sources, such as airport and port operations, and the formation of nucleated particle (NP) (Blangiardo et al., 2013; Clifford et al., 2012; González et al., 2011; Imhof et al., 2005; Kumar et al., 2008; Mazaheri et al., 2013a; Morawska et al., 2008; Seigneur, 2009; Westerdahl et al., 2008; Zhu et al., 2002). Meteorological parameters, in particular wind speed and its direction, have a significant impact on air pollution dynamics and the process of NP formation (Elminir, 2005; Zhao et al., 2015). While primary urban particles are emitted into the atmosphere by various sources, NPs are formed by complex reactions in the atmosphere, of which traffic-related emissions are considered their major precursors (Dall'Osto et al., 2013). There are several NP formation mechanisms, such as binary nucleation (H_2SO_4 and water vapour), ternary nucleation (NH_3, H_2SO_4, and water vapour) and ion-induced nucleation for charged particles (Kulmala et al., 2004). While existing nucleation theories are unable to explain

* Corresponding author.
E-mail address: l.morawska@qut.edu.au (L. Morawska).

http://dx.doi.org/10.1016/j.atmosres.2017.04.036
Received 14 September 2016; Received in revised form 26 April 2017; Accepted 28 April 2017
Available online 29 April 2017

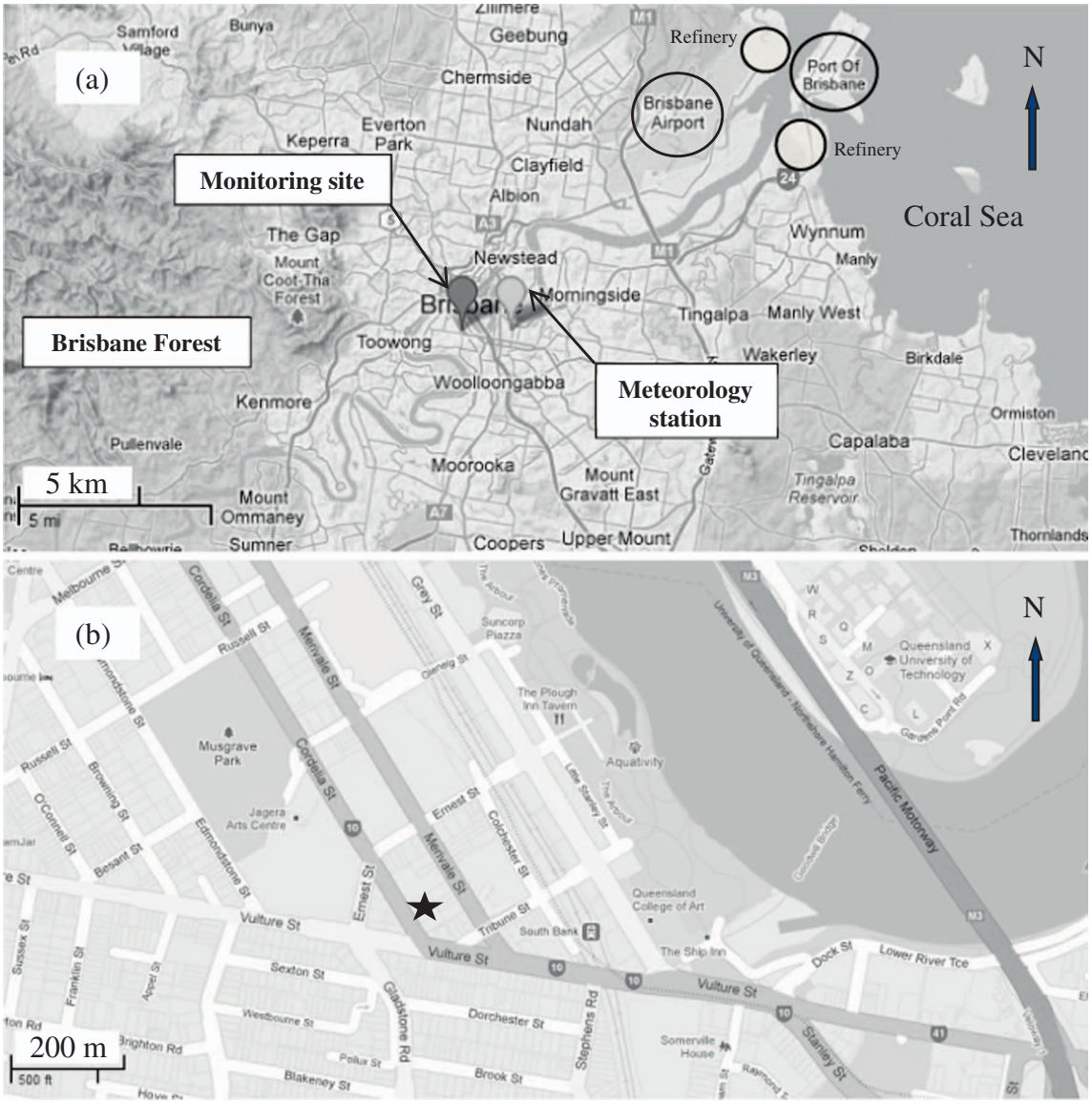

Fig. 1. Map of the sampling location in two different scales: (a) the monitoring site and meteorological data collection site of BOM; (b) the air quality monitoring site (marked by a star) adjacent to the streets.

NP formation mechanisms in many circumstances, recent investigations have confirmed that, along with H_2SO_4 vapour, atmospheric low volatile organic compounds might play a significant role in NP formation events (Kerminen et al., 2010; Paasonen et al., 2010; Wang et al., 2015). A large-scale chamber experiment, CLOUD, results suggest that the time-resolved H_2SO_4 and ammonia concentrations in a typical boundary layer are too low ($^{<}$1 ppb) to account for observed NP (Kirkby et al., 2011).

NPs were also observed during the highest solar radiation period of the day, when the pre-existing particle concentrations were relatively low (Cheung et al., 2011a; Jung et al., 2013; Wood, 2008; Wood, 2011). Noon-time NP formation has been widely observed in many urban areas (e.g., Beijing, China (Donaldson et al., 2001), Singapore (Betha et al., 2013), Brisbane, Australia (Cheung et al., 2011b), and Sapporo, Japan (Jung et al., 2013)). Reche et al. (2011) found that increase in midday PNC in European urban background areas is linked to NP formation events. Depending on the meteorology and characteristics of available NP formation precursors in the ambient air, the noon-time NP growth rate varied significantly in urban locations. For example, Wu et al. (2007) found a NP growth rate of 0.1 to 11.2 nm/h in urban areas of Beijing, China, which is comparable to those found in the Brisbane urban airshed (1.79 to 7.78 nm/h) (Cheung et al., 2011b). Formation of NPs in ambient air results in a quick rise in overall particle number concentrations of one to two orders of magnitude, depending on the existing pollution sources and meteorological factors in the investigated area (Morawska et al., 2008).

K-means clustering algorithm was applied in urban locations in Europe and Australia, to apportion the major urban sources contribution to UFPs (Brines et al., 2015; Salimi et al., 2014). However, the k-means clustering algorithms are unable to quantify day to day variations, and its results can be uncertain due to its unsupervised data classification method (Boutsidis et al., 2009; Leiva and Vidal, 2013). Therefore, the application of clustering algorithms to isolate atmospheric source contribution to UPFs can be imprecise for dynamic conditions. In addition, it has been shown that only relying on mathematical modelling results in an underestimation of the mass concentration of secondary organic compounds in urban areas (Volkamer et al., 2006). Due to the limitation of modelling approaches, the contribution of NP to UFPs is likely to be underestimated in urban areas.

Bayesian framework has emerged as a promising mathematical modelling tool for environmental systems because of its robustness over