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1	The influence of wind speed on new particle formation events in an urban
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22 Abstract

New particle formation (NPF) is a common occurrence in urban environments where it is promoted by a ready supply of gaseous precursors such as sulphuric acid, ammonium and volatile organic compounds originating mainly from motor vehicle emissions. In this paper, we investigate the influence of wind speed on the occurrence of NPF events in a subtropical urban environment. The analysis was based on a large data set obtained with a neutral cluster and air ion spectrometer (NAIS) over 485 days of observations, out of which NPF events were observed on 213 days. Particles formation was most likely to occur during the morning hours and we observed NPF events between 8 am and noon on 123 of these days. In order to assess its influence, we estimated the mean wind speed between 8 and 9 am on these 123 days and on the days with no NPF. We found a statistically significant difference in average wind speeds during days with NPF (1.31 m s⁻¹) and days with no NPF (1.84 m s⁻¹) (p<0.05). Several studies have shown that increasing wind speed enhances NPF in polluted megacities by removing pre-existing particles. Our observations indicate that this effect is less important in smaller cities, so that higher wind speeds can mitigate NPF by the removal of condensable gases. The wind speed does not affect the rate of NPF in clean environments.

Keywords: New particle formation, wind speed, atmospheric aerosols, secondary particles.

1 Introduction

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New particle formation (NPF) occurs through homogeneous nucleation and is one of the major sources of particles in the atmosphere. The process requires a condensable precursor gaseous species that, in urban environments, are mainly sulfuric acid and semi-volatile organic compounds emitted by motor vehicles and industrial facilities (Kulmala et al., 2013). The concentration of gaseous precursors in a given environment is controlled by a number of factors including the type and intensity of the sources. Pre-existing aerosols act as a condensation sink to gases resulting in a reduction in their vapour pressure and may inhibit homogeneous nucleation and the occurrence of NPF (Seinfeld and Pandis, 2006). Thus, there is a trade-off between the concentrations of precursor gases and particle pollution in a given environment and this balance is also affected by meteorological parameters such as solar radiation, temperature, relative humidity, wind direction and wind speed (Birmili and Wiedensohler, 2000). Many of the characteristics of NPF events in the urban environment of Brisbane, Australia, such as their frequency and times of occurrence, formation and growth rates and relationships to gaseous concentrations in the air, have been extensively studied and reported in the literature (Cheung et al., 2011; Crilley et al., 2014; Jayaratne et al., 2016; Pushpawela et al., 2018). In this study, we focus on the effect of wind speed on the frequency of occurrence of NPF events. This

is an aspect that has not been studied in great detail anywhere in the world, and there is very

little information available in the literature. Wind is important in polluted cities where it serves

to remove particles from the air and a limited number of studies in such locations have noted

an increase in the frequency of occurrence of NPF events when the wind speed is higher 65

(Hamed et al., 2007; Zhang et al., 2011; Wang et al., 2013; Zhang et al., 2017).

However, at a location close to a pollution source, wind can also remove particles and gaseous precursors, leading to an imbalance in the conditions favourable for NPF. This study was carried out in order to investigate the effect of wind speed on the frequency of NPF events in Brisbane, Australia, which is a medium-sized city dominated by an urban environment. It was facilitated by a large dataset of particle number concentrations obtained on nearly 500 days, spanning a period of five years, using a neutral cluster and air ion spectrometer (NAIS). The NAIS can measure particles down to 2 nm in size, which is where the initial steps of nucleation and formation of particles occur (Manninen et al., 2011;Manninen et al., 2016). Thus, it has the ability to identify NPF events and differentiate them from particle growth events that take place with no NPF (Pushpawela 2018a). We analysed the data during periods when there were NPF events and compared them with periods when there were no NPF events and observed a statistically significant difference in wind speeds between the two periods. We compare this result with observations of wind speeds during NPF events that have been reported in the literature at other locations around the world and draw some interesting conclusions.

2 Methods

The NAIS was installed on the top floor of a six-floor building at the Queensland University of Technology in Brisbane, Australia, where it sampled the air from outside a window. The University campus is situated on the banks of the Brisbane River, approximately 100 m away from a busy motorway carrying about 120,000 vehicles per day. The site is less than 1 km away from the Central Business District of Brisbane and is representative of a typical urban environment in Australia. The measurements were carried out between 2012 and 2017, yielding a large data set of 485 complete days that was used in this analysis. The NAIS was unavailable

on the other days mainly as it was required in related projects at other locations and also due to various reasons such as instrument failure, routine cleaning and service.

Considering the proximity to the motorway and the city centre, the pollutants at this site were mainly from motor vehicle exhaust emissions. Emissions may also be received from the Port of Brisbane and two oil refineries as well as from Brisbane Airport, all located about 20 km away from the monitoring site. Meteorological data including wind direction and wind speed were obtained from the Department of Environmental and Heritage Protection, Queensland, air quality monitoring station which was located on the roof of the same building.

The NAIS is manufactured by Airel Ltd, Estonia, and is specifically designed to detect NPF events (Manninen et al., 2016). It measures the size distribution of charged and neutral particles in the size range from 2.0 - 42 nm. The instrument has a high-resolution time down to 1 s and consists of two cylindrical electrical mobility analysers, one for each polarity. It cycles between two operational modes of 2 min each: the ion mode and the particle mode. In the ion mode, the NAIS measures naturally charged particles without any modification. In the particle mode, it measures all charged and uncharged particles. The lower detection limit in the particle mode is restricted to 2 nm due to presence of corona generated ions in the instrument (Manninen et al., 2016). A more detailed discussion of its design and principles may be found in (Manninen et al., 2011) and (Mirme and Mirme, 2013).

NPF events were identified using the rate of change of total particle concentration, dN/dt, where N is the number of particles in the size range 2-10 nm and using the classification given by (Zhang et al., 2004). Events with N > 5000 cm⁻³ for at least 1 hour and dN/dt > 5000 cm⁻³ h⁻

were defined as NPF events. All of these events started in the nucleation mode size range and prevailed over a time span of more than one hour, generally exhibiting a "banana" shape in the time-series contour plot of particle number concentration (PNC), indicating particle formation and subsequent growth. We considered only those NPF events that began between 8 am and noon. We estimated the mean wind speed between 8 and 9 am on these 123 days and on the 272 days with no NPF events at all. Statistical significances of the difference between the mean wind speeds on the two sets of days were calculated using the Student's t test with a confidence interval of 95%. In order to assess the influence of wind speed, we calculated the ratio of the wind speed on NPF days to that on non-NPF days and defined this as the 'wind speed ratio'.

3. Results and Discussion

3.1 Observation of NPF during the study period

NPF events were observed on 213 of the 485 days on which we were able to obtain data. Figure 1 shows a typical spectragram of an NPF event as measured by the NAIS. We see that the NPF began at around 9.30 am and the mean particle size reached 10 nm at about 11.30 am, giving a mean particle growth rate of 5 nm h⁻¹, which is typical for NPF events in Brisbane (Pushpawela et al., 2018). The total particle number concentration observed by the NAIS increased from 4.5×10^4 cm⁻³ to 8.7×10^4 cm⁻³ during these two hours. The corresponding number concentrations of particles in the size range 2-10 nm were 2.3×10^4 cm⁻³ to 5.9×10^4 cm⁻³, giving a dN/dt value of 1.8×10^4 , indicating that this was a relatively strong NPF event.

Most of the NPF events occurred during the morning hours, with a peak frequency between 8 and 10 am. Out of the 213 days, NPF events were observed between 8 am and noon on a total

of 123 days. This is likely to be a result of several factors such as the higher concentration of precursor gases from motor vehicles during the morning rush hour and the onset of solar radiation. Solar radiation intensity was an important parameter that had a strong influence on the likelihood of an NPF event. In general, NPF events were less likely to occur on cloudy days with low solar irradiance. The mean solar radiation intensity between 8 and 9 am on NPF days were significantly higher than that on the days with no NPF, their mean values being 505 W m⁻² and 397 W m⁻², respectively. While NPF events were more likely to occur on days with lower relative humidity (Pushpawela 2018a), we did not observe similar differences in air temperature or wind direction.

Interestingly, in our earlier study (Pushpawela 2018a) we observed no difference in average wind speeds between NPF days and non-NPF days. This conclusion was based on mean wind speeds between 8 and 9 am and NPF events that occurred at all times of the day. In the present study, the analysis consider only those 123 days when an NPF event occurred between 8 am and noon, alongside the mean wind speeds between 8 and 9 am. Now, we observed a clear statistically significant difference between the corresponding average wind speeds on NPF days and days with no NPF. Figure 2 shows the number in each of these two sets of days as a function of wind speed binned into intervals of 0.25 m s⁻¹. The mean wind speeds between 8 and 9 am on days with an NPF event between 8 am and noon and days with no NPF events at all were 1.31 m s⁻¹ and 1.84 m s⁻¹, respectively, giving a wind speed ratio of 0.71. The difference between the two means was statistically significant at a confidence level of 95%.

Next, we look at how this result compares with observations at other locations. Although, the characteristics of, and conditions giving rise to, NPF events have been extensively studied

and reported, there are no studies that have specifically investigated the effect of wind speed on NPF. Moreover, the number of publications that have provided wind speeds during and outside NPF events is highly limited. Table 1 gives a summary of the papers that have provided this information. We have separated the studies according to the level of pollution into clean sites (3 studies – a to c) and widely polluted sites (5 sites – d to h). As a measure of the relative effect of wind speed, we derived the ratio of the wind speed reported during NPF events to the wind speed reported during other times. It must be stated that, in some cases, the wind speed is specified at the time of an event and in others, it is merely the average wind speed over a complete day, so the analysis may be affected to some degree. Despite these shortcomings, there is a clear difference in the value of the ratio between the clean sites and the widely polluted sites (Figure 3). The wind speed ratio at the clean sites were very close to 1.0, indicating that the wind speed had very little impact on the occurrence of NPF events. This makes sense as the precursor gases at clean sites are generally volatile organic compounds from vegetation or maritime sources that are widely distributed, such that their concentrations are spatially constant. Winds are less likely to affect the precursor concentrations at such sites that may include clean forest, mountain or coastal locations. On the other hand, winds have a strong effect on the pollution levels in large cities and widely industrialised regions. Several studies in Beijing have reported the near absence of NPF events during heavy pollution events (Wehner et al., 2004; Wu et al., 2007; Guo et al., 2014; Jayaratne et al., 2017). This is because the gaseous precursors condense on the preexisting particles, reducing supersaturation and inhibiting NPF (Kulmala et al., 2016). Clean winds from cleaner regions outside sweep the airborne particles away, improving the air quality in the city and providing conditions favourable for NPF. This explains why the wind speed ratio is significantly greater than 1.0 in the studies carried out at the five widely polluted sites.

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In the present study in Brisbane, the wind speed ratio was 0.71, which is significantly less than the values reported from the large cities listed in Table 1 and Figure 3. In comparison to major cities around the world, Brisbane is not a polluted city. The mean annual PM_{2.5} concentration in the city is below 10 µg m⁻³. In comparison to the five locations d to h, Brisbane may be considered a 'clean' location. The proximity of the site to the nearby freeway will play a role in the background precursor gas concentration. Previous studies at this site have shown that NPF occurs on about 40% of days and the gaseous precursors originate from motor vehicle emissions (Cheung et al., 2011;Crilley et al., 2014;Pushpawela et al., 2018). These gases tend to accumulate at the site during calm weather, promoting conditions for NPF, and disperse when the wind speed increases. This may explain the wind speed ratio of less than 1. One other study that was carried out in a medium-sized city and reported wind speed values during NPF events was (Cheung et al., 2013) in Taipei, Taiwan. The wind speed ratio at this location was 0.81, which is close to the Brisbane value.

4. Conclusions

We conclude that the effect of wind speed on NPF in the atmosphere depends on the distribution of pollutants around the location. In clean environments such as forests, mountainous and coastal regions, wind speed has little effect on the likelihood of NPF events. This is because the precursor gas concentrations are uniformly distributed and are not affected by the wind. In large cities, NPF can be inhibited during stagnant conditions when pre-existing particles suppress homogeneous condensation in the atmosphere. Cleaner winds from outside the city generally serve to remove the particles, promoting NPF. Thus, in widely polluted regions, the likelihood of NPF will increase with wind speed. This has been observed in megacities such as Beijing and in large industrialised regions such as the Pearl

River Delta in China, and the Po Valley in Italy. In a 'clean' city like Brisbane, the particle concentration in the air is low and, therefore, removal of particles by the wind does not make a large difference to the condensation sink. However, together with the particles, the wind will remove precursor gases that are produced by local sources, inhibiting NPF. This will lead to a wind speed ratio that is less than 1. Therefore, in summary, in larger more polluted cities, higher wind speeds will enhance the chances of NPF, whereas in smaller cities with less pollution, higher wind speeds can mitigate the chances of NPF.

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321	Figure captions
322	Figure 1: NAIS spectragram of a typical NPF event in Brisbane
323	Figure 2: Number of NPF days and non NPF days against the mean wind speed between 8
324	and 9 am on that day.
325	Figure 3: The wind speed ratio at various sites derived from the literature. Details are in
326	Table 1
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346 Figures

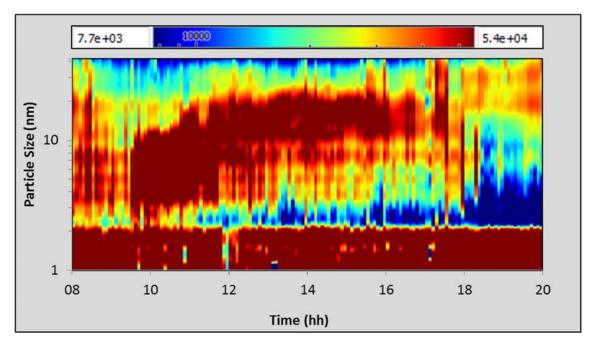


Figure 1: NAIS spectragram of a typical NPF event in Brisbane.

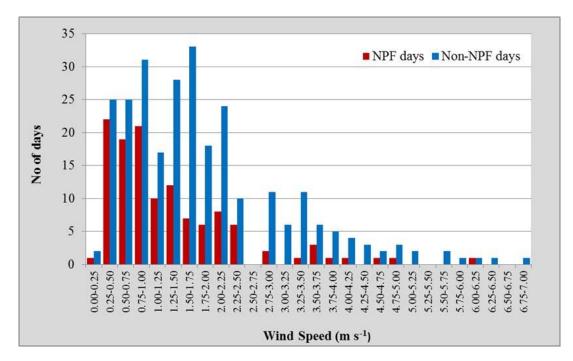


Figure 2: Number of NPF days and non NPF days against the mean wind speed between 8 and 9 am on that day.

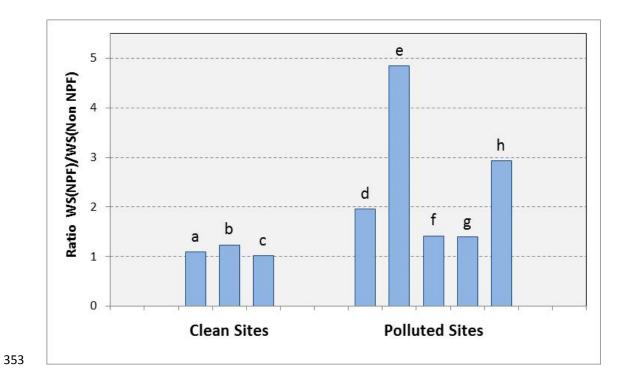


Figure 3: The wind speed ratio at various sites derived from the literature. Details are in Table 1.

Table 1: Summary of previous studies that have investigated the effect of wind speed on NPF events. The data labels refer to Figure 3.

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Data	Study	G.	Mean wind speed (m s ⁻¹)		Wind Speed	
Label in Fig 3		Site -	NPF events	Non events	Ratio	Comments
			Clean Si	tes		
a	Lunden et al. (2006)	Nevada CA, USA	1.91	1.75	1.09	Clean mountain site
b	Charron et al. (2007)	Harwell, UK	5.7	4.6	1.24	Clean rural site
С	Zhang et al. (2017)	Anhui Province China	5.1	5.0	1.02	Clean mountain summit
Widely Polluted Sites						
d	Hamed et al. (2007)	Bologna, Po Valley, Italy	4.5	2.3	1.96	Industrial area with high population density
e	Guo et al. (2012)	Hong Kong	1.98	0.41	4.84	Mountain overlooking major city
f	Wang et al. (2013)	Pearl River Delta, China	2.7	1.9	1.42	Rural/built up area
g	An et al. (2015)	Nanjing, China	2.8	2.0	1.40	University campus in major city,
h	Jayaratne et al. (2017)	Beijing, China	2.4	0.82	2.93	Polluted city
		M	edium-size	d Cities		
	Cheung et al. (2013)	Taipei, Taiwan	2.61	3.23	0.81	Urban site
	Present Study	Brisbane, Australia	1.31	1.84	0.71	Urban site