The effects of vehicle emissions and nucleation events on vertical particle concentration profiles around urban office buildings

T. N. Quang, C. He, L. Morawska, L. D. Knibbs, and M. Falk

Responses for ACPD Referee #2's Comments

Received and published: 16 March 2012

The authors thank the Referee for his/her constructive and useful comments.

General Comments:

Comment G1

- The title is not suitable for this study because there was not enough evidence how traffic emissions or new particle formation would influence in the vertical profiles. In my point of view, the title is better written in the form "Vertical profiles of fine particle number size distributions and PM2.5 concentration around three buildings in the urban area of Brisbane, Australia"

Response G1

The title has been revised to "Vertical profiles of particle concentration around urban office buildings".

Comment G2

- The introduction and the literature review should contain most of the relevant studies regarding vertical profiles of fine aerosols in urban areas. In that sense, the reference list should be longer and more extensive.

Response G2

To address the above, more related studies have been added to the research background and included in a new paragraph which has been added to the end of page 1616, as follows.

'In addition to research surrounding building envelopes, some studies have quantified the vertical profiles of particle concentrations in urban areas. Imhof et al.(2005) has shown that PN concentrations 60 m downwind of a highway decreased when measured at heights of 5 – 30 m. Zhu and Hinds (2005) quantified the vertical particle concentrations measured 50 m downwind of an elevated highway and reported that the PN concentrations increased within the first 5m from the ground, then decreased at higher levels. He and Dhaniyala (2012) measured vertical profiles of PN concentrations at heights between 0.55 to 10 m at distances 15, 50, and 100 m from a highway. Their results have shown that vertical profiles of particle concentrations vary with wind speed, direction and distance from the highway.'

Comment G3

- While I emphasize it here as "fine particles", authors did not. I believe that they should in the first place consider this fact because their instrumentation provided a certain fraction of fine particles (size distribution between 8.5 – 400 nm) in addition to the PM2.5. Although, the authors have to discuss the validity of their size distribution for the whole fine particle size range (say 3-1000 nm).

Response G3
As reported in section 2.2 of the manuscript, we not only measured the PNSD in the size range of 8.5 – 400 nm and $PM_{2.5}$ concentration, but also the PN concentration in the size range of 6 – 3000 nm. To address the Reviewer's comment above, the PN concentrations in different fractional sizes have been added and discussed further in sections 3.3 and 3.4.

Comment G4
- Another important terminology error in this article is that authors mentioned in several places the term "PNSD concentrations" denoting for Particle Number Size Distribution concentration. The correct term should be "particle number size distribution". [Please mind this typo]

Response G4
The term "particle number size distribution concentration" has been revised to *"particle number size distribution (PNSD)"*.

Comment G5
- While trying to make evidence of traffic emissions influence, the authors has to present traffic activities data and make real statistical analysis for it with respect to the aerosol data. Otherwise, the presentation of the results would be like hand waving and without real support. Another important point here regarding traffic is to consider workdays versus weekends.

Response G5
The traffic flow on the streets close to Buildings A, B and C have been added to the relevant figures, respectively. Please refer to the response S2.3 to Referee 1. Traffic flows close to Buildings B and C were not monitored simultaneously with aerosol data, but gathered from the Queensland traffic and travel information website (http://131940.qld.gov.au/Traffic-Census.aspx). Therefore, we did not conduct any statistical analysis with particle concentration data at these building sites due to the lack of contemporaneous traffic data.

Comment G6
- To make a good presentation for then nucleation events, I strongly recommend quantifications of the nucleation and growth rates of the events. Furthermore, when the authors presented the differences in the vertical profiles during nucleation events and compared it with the daily ones, did they consider all the data in the daily averages or just removed the time periods of nucleation events? Please specify what does "daily" state for specifically.

Comment S3
- Section "2.5. Identification of nucleation events": I strongly recommend the differentiation between local events and regional events in the urban atmosphere. For example, the original criteria by Dal Maso (2005) and the modified one by Hussein et al. (2008) described the regional events. However, Hussein et al. (Atmos. Chem. Phys., 9, 4699-4716, 2009) presented how to distinguish local events from regional ones. The main key factor is the time-span of the event.

Response G6 and S3

The issues raised by the reviewer are addressed in the response G1 to Referee 1.

The "daily" aspect refers to the 24-hour average on days characterised by the occurrence of a nucleation event.

Comment G7

- It is very interesting to compare the vertical profiles found in this study to those found in open lands nearby traffic. there are couple of interesting studies in the literature dealing with this topic.

Response G7

A few related studies on open land particle concentration vertical profiles have been added to the research background, and we agree that it is an interesting point of comparison. However, we feel that to compare our results/findings with these studies is outside the scope of this work, as the measurement conditions (such as distance to the highway, wind speed, wind direction ...) were not analogous.

Comment G8

- Please consider specific size fractions as well in addition to the total number concentration and PM2.5.

Comment S9

- Figure 10: you should merge subfigures a and b together. also merge subfigure c and d. Please do the same for other similar figures.

Response G8 and S9

The PN concentrations in specific fractional size ranges have been added and the text and figures in section 3.3 have been revised as follows (highlighted words have been newly added or modified).

3.3 Vertical profiles of particle concentrations

The average vertical profiles of the PNSD and $PM_{2.5}$ for the entire day, rush-hours and during nucleation events at Buildings A, B, and C are presented in Figs. 10, 11 and 12, respectively (these figures are shown in the appendices). It should be noted that the data of the nucleation events at Building C were only collected at rooftop and street levels and therefore, constructing a vertical profile based on nucleation events at this building, was not appropriate. However, the measured results at Building C show that the PN concentration at rooftop levels was significantly higher than at street levels during the event, while the opposite was the case for the $PM_{2.5}$ concentration.

At Building A, the trends of TNC and $N_{<30}$ were similar. Their average concentrations during nucleation events themselves and over 24-hour on the day of nucleation events constantly increased with height ($p < 0.01$). While during the rush-hours, they decreased between 1.5 and 10.5 m, and then increased onward ($p < 0.05$). In contrast, the trends of $N_{30\text{-}100}$ and $N_{>100}$ fluctuated and depended on the measurement heights and times. In general, the average daily $PM_{2.5}$ concentrations decreased with increasing height, however they stabilised at heights between 6.5 and 10.5 m. During rush-hours, $PM_{2.5}$ concentrations were higher at heights of 6.5 and 10.5 m, but lower at a height of 14.5 m, compared to the daily concentrations ($p < 0.05$). The $PM_{2.5}$ concentrations during the nucleation events were generally lower than the daily concentrations ($p < 0.01$).

At Building B, $N_{30\text{-}100}$, $N_{>100}$ and $PM_{2.5}$ concentration at street levels were always higher than those at rooftop levels ($p < 0.05$). During the rush-hour periods, TNC and daily average concentration was significantly higher at street level than at rooftop level, but the opposite was the case during the nucleation events ($p < 0.05$). $N_{<30}$ at rooftop level was significantly higher than at street level during the nucleation event ($p < 0.01$), while their daily and rush-hour concentrations were relatively similar (p value of 0.17 and 0.78, respectively).

The average daily PNSD and $PM_{2.5}$ concentration decreased with height between 1.5 and 21.5 m at the rear (opposite side facing the road) of Building C ($p < 0.01$), however N_{30-100}, $N_{>100}$, $PM_{2.5}$ tended to stabilise at heights between 5.5 and 9.5 m, followed by a less pronounced decrease from 9.5 to 21.5 m. During the rush-hour periods, N_{30-100}, $N_{>100}$, TNC decreased from 1.5 to 9.5 m, and then stabilised at heights between 9.5 and 21.5 m. $N_{<30}$ increased at the beginning of the rush-hour period, then decreased from 5.5 to 9.5 m, and finally stabilised onwards. The rush-hour $PM_{2.5}$ followed the $PM_{2.5}$ daily trends and was higher than the daily concentrations.

In general, the trend of TNC followed those of $N_{<30}$ or N_{30-100} during the nucleation event and rush-hours, respectively, while the trends of $N_{>100}$ and $PM_{2.5}$ were similar.

At Building B, the daily and rush-hour PN concentrations at street level were higher than those on the rooftop. This finding is in agreement with the results of previous studies (Hitchins et al., 2002; Kumar et al., 2009; Li et al., 2007; Longley et al., 2004; Väkevä et al., 1999). On the contrary, the daily and rush-hour PN concentrations at Building A increased with height. This is likely to be attributed to the fact that the busway is located close to the building and elevated above ground level, and therefore, it has a stronger influence on the concentrations measured at higher levels compared to Building B. The daily and rush-hour PN concentrations at the rear of Building C decreased with increasing height. This finding is not in agreement with the results reported by Hitchins et al. (2002) based on measurements in Brisbane, where a short time measurement (5 samples during 450 s for each level) was conducted. The difference could be due to the highly diurnal variations of influencing factors, such as vehicle emissions, wind speed and wind direction on particle concentrations between the different levels of this building.

The $PM_{2.5}$ concentrations seemed to consistently decrease with height throughout the day and this finding is also in accordance with previous research (Chan and Kwok, 2000; Horvath et al., 1988; Micallef and Colls, 1998; Rubino et al., 1998). However, the $PM_{2.5}$ concentrations at Buildings A and C did not decrease consistently. In the case of the Building A, this may be due to the influence of the proximity of the busway. The sampling points were located on the rear side of Building C and were obstructed by other buildings located behind it, and therefore, some stagnation of air in this region may have influenced the $sPM_{2.5}$ concentrations at mid-height levels.

In general, the vertical profiles of the $PM_{2.5}$ concentrations around the building envelopes decreased with increasing height. However, vertical profiles of the PNSD were building-specific and the rate of change with height was different at all three buildings. The results indicate that it is not only vehicle emissions that influence the particle vertical profiles, but new particle formation as well; while particle number increased, we observed a reduction in particle mass during the nucleation events. These results serve to further define the specific effect of roadway proximity and nucleation formation on the vertical profiles of PN and $PM_{2.5}$ concentrations around building envelopes. Moreover, the highly building-specific nature of the profiles and factors affecting them underscores that, ideally, measurements form the basis of any modelling or planning exercise prior to or after construction of a building. Such an approach, which is currently lacking for the most part, will ensure the greatest model veracity. This has important implications for selecting appropriate sites for the air intakes of building HVAC systems to minimise occupant exposure to combustion products, and also to investigate how street-level exposures may be mitigated via improved design practices.

Specific comments:

Comment S1

- Section “2.1. Setting”: I suggest making a map showing the locations of the three sites within the urban Brisbane. This is to give a better insight into the distribution of sources

around each building. It is not enough to have the closest sources, but also further ones as well.

Response S1
A map showing the locations of the three sites in Brisbane has been added to section 2.1 (see Fig. 1 in the appendices).

Comment S2
- Section "2.2. Instrumentation": a detailed description of the sampling lines, losses, and calibration. This might explain why the nucleation events were observed only at the rooftop and street levels at Building C.

Comment S4
- Section "3.3. Vertical profiles of particle concentrations": how come the nucleation events were observed at the street level and on rooftop of building C only and they were not observed at the middle levels?!

Comment S8
- Figure 6: plot other levels in addition to the rooftop and street level. The same apply for similar figures.

Response S2, S4, and S8
As described in section 2.3, we used two sets of instruments; one measuring continuously at the reference site (at the highest level) for each building, and the other measuring simultaneously at a lower level. In the case of Building C, one set of instruments sampled continuously at the reference site, which was located 21.5 m above the ground, and 13.5 m above and 7 m away from the freeway. The second set was moved between sites located at heights of ~1.5 m, 5.5 m, 9.5 m and 21.5 m (levels 1, 2, 3 and 6, respectively) on the opposite side of the building (the rear of the building). The sampling sites and building layout are shown in Fig. 3 (Figure 4 – new) of the manuscript.

We observed 3 nucleation events during the measurements at Building C (refer to section 3.2.2 for more detail). Two events occurred when the second set of instruments was measuring at the rear of level 3 (street level), and one occurred when the second set was measuring at the rear of level 6. No nucleation event was identified when the second set was measuring at the rear of levels 1 and 2.

However to clarify the effect of nucleation events on the particle vertical profiles at Building C further, lines 2-3, page 1628, in section 3.3 has been revised from "noting that ... street levels" to *". It should be noted that the data of the nucleation events at Building C were only collected at rooftop and street levels"*.

Fig. 6 presented the PNSD spectra at Building A, on 7 August 2009, when two sets of instruments measured at the rooftop and street levels only.

This approach was also used at Buildings B and C.

Comment S5
- Page 1630, lines 21-22: Unclear statement "This suggests a more pronounced influence from vehicle emissions and new particle formation on PN and PM2.5 concentrations at each level during these periods."

Comment S10
- Figure 13: This figure is very interesting and it is the main finding of the study. Please make extensive discussion about it. Also do the same discussion regarding the different particle size fractions.

Response S5 and S10
Relationships between the PN size fractions and $PM_{2.5}$ concentration at different levels and buildings have been added and the text, figures and table in section 3.4 have been revised as below (highlighted words have been newly added or modified).

3.4 Relationship between PNSD and $PM_{2.5}$ concentration

Spearman's correlation coefficients (rho) for the PNSD and $PM_{2.5}$ concentrations at different heights and different time periods at Buildings A, B and C are presented in Figs. 13, 14, 15, respectively, and Table S2 (see these figures and table in the appendices). However, as noted, new particle formation data was collected only at the reference site and street level during the measurement campaign of Building C. Therefore, correlations between the PNSD and $PM_{2.5}$ during the nucleation events at this site were not calculated. In general, the correlation coefficients between $N_{>100}$ and $PM_{2.5}$ were higher, while the correlation coefficients of $N_{<30}$ were usually lower compared to other particle size fractions.

The PNSD and $PM_{2.5}$ correlation coefficients on the rooftop were higher than those at street level at Building B. The difference between correlation coefficients for PN size fractions and $PM_{2.5}$ concentrations at Building A were higher than at Building B. This is likely due to the relative proximity of the particle sources at each level, as well as to the closeness to the busway at Building A. Both daily and rush-hour correlation coefficients of PNSD at the rear of Building C initially increased from the ground to level 3, and then decreased closer to the rooftop.

Correlations between the PNSD and $PM_{2.5}$ were characterised by a significant variability and dependence on particle size fraction, measured height and particle emission sources. The linear correlations for the building envelopes, especially during the rush-hour and nucleation events, fluctuated significantly. This indicates that it is not appropriate to use particle mass concentrations to infer PN concentrations when modelling vertical concentrations around the building envelope and at a street level. This finding, while not a novel observation, adds weight to the existing case for separately considering particle mass and number during any urban modelling or exposure assessment exercise.

Comment S6
- Page 1631, lines 14-15: unclear statement "In general, vertical profiles of PM2.5 concentrations around the building envelopes were markedly higher with decreasing distance to nearby streets."

Response S6
To clarify, lines 14-15, page 1631, section 4, the sentence 'In general ... nearby streets' has been revised to *'In general, vertical profiles of $PM_{2.5}$ concentrations around building envelopes showed a consistent decrease in concentration with increasing distance from nearby streets'*.

Comment S7
- Figure 4: It is more interesting to plot longer time periods. Also the same comment for similar figures in the supplementary files.

Response S7
Figure 4 (Figure 5 – new) illustrates the daily average fractional size particle concentrations, based on the entire sampling campaign at each building (2-3 weeks).

Comment S11
- When plotting the size distribution spectra, please use logarithmic colour scale instead of linear. That would make better visibility of other interesting events in the size distributions.

Response S11
The logarithmic colour scale has been used in the size distribution spectra.

References

He, M., and Dhaniyala, S.: Vertical and horizontal concentration distributions of ultrafine particles near a highway, Atmospheric Environment, 46, 225-236, 2012.

Imhof, D., Weingartner, E., Vogt, U., Dreiseidler, A., Rosenbohm, E., Scheer, V., Vogt, R., Nielsen, O. J., Kurtenbach, R., Corsmeier, U., Kohler, M., and Baltensperger, U.: Vertical distribution of aerosol particles and NOx close to a motorway, Atmospheric Environment, 39, 5710-5721, 2005.

Zhu, Y., and Hinds, W. C.: Predicting particle number concentrations near a highway based on vertical concentration profile, Atmospheric Environment, 39, 1557-1566, 2005.

Appendices

Table S2. Spearman's correlation coefficients (rho) for PNSD and $PM_{2.5}$ concentration around the building envelopes

Site	Measured height	Time period	Spearman's correlation coefficient (rho)			
			$N_{<30}$	N_{30-100}	$N_{>100}$	TNC
Building A	1.5 m	Daily	0.05	0.68**	0.80**	0.63**
		Rush-hours	0.21	0.22	0.78**	0.24
		Nucleation	0.49	0.63*	0.66*	0.48
	6.5 m	Daily	0.04	0.85**	0.94**	0.67**
		Rush-hours	0.46*	0.66**	0.52**	0.56**
		Nucleation	0.26	0.69**	0.71**	0.69**
	10.5 m	Daily	-0.20*	0.72**	0.88**	0.29**
		Rush-hours	0.12	0.77**	0.80**	0.49**
		Nucleation	0.17	0.72**	0.36	0.18
	14.5 m	Daily	-0.11	0.84**	0.96**	0.43**
		Rush-hours	0.27	0.60**	0.67**	0.51**
		Nucleation	-0.03	0.73**	0.90**	0.39*
Building B	1.5 m	Daily	0.53**	0.69**	0.82**	0.72**
		Rush-hours	0.13	0.20	0.64**	0.38
		Nucleation	0.66**	0.65**	0.57**	0.65**
	78.5 m	Daily	0.69**	0.82**	0.89**	0.84**
		Rush-hours	0.22	0.35	0.76**	0.43*
		Nucleation	0.78**	0.85**	0.87**	0.87**
Building C	1.5 m	Daily	0.50**	0.40**	0.44**	0.45**
		Rush-hours	0.46*	0.33	0.5*	0.41*
	5.5 m	Daily	0.37*	0.74**	0.75**	0.68**
		Rush-hours	0.55**	0.57**	0.82**	0.61**
	9.5 m	Daily	0.40*	0.85**	0.9**	0.79**
		Rush-hours	0.62**	0.68**	0.68**	0.69**
	21.5 m	Daily	0.56**	0.79**	0.60**	0.74**
		Rush-hours	0.31	0.44*	0.38*	0.46*

*. Correlation is significant at the 0.05 level (2-tailed)

**. Correlation is significant at the 0.01 level (2-tailed)

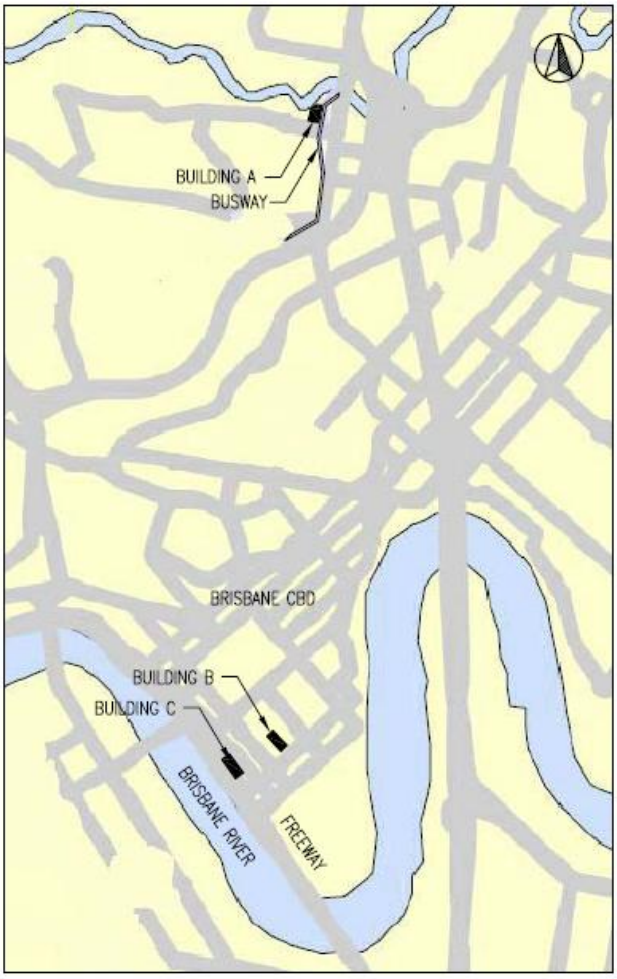

Fig. 1. Locations of Buildings A, B, and C.

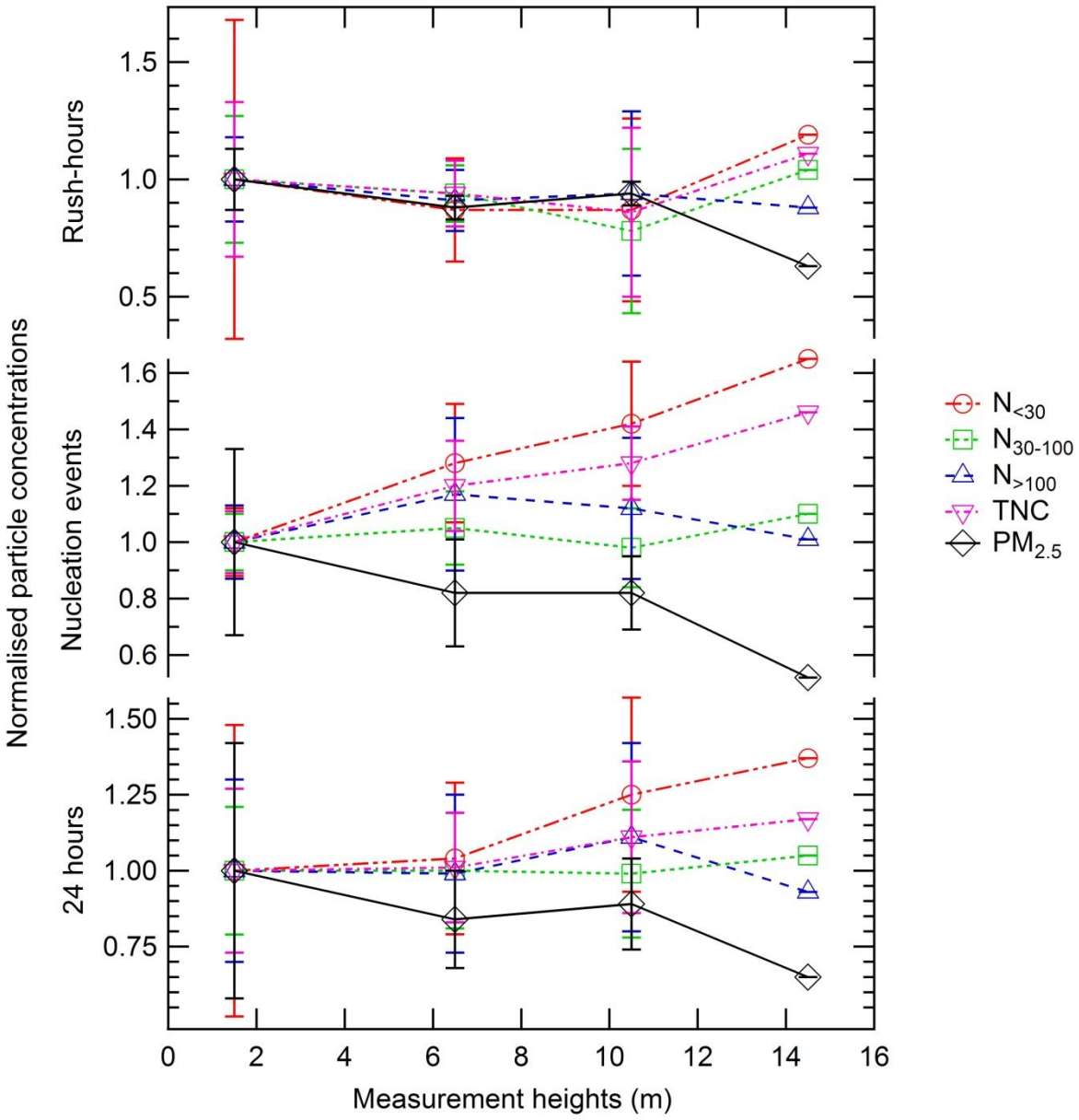

Fig. 10. Vertical profiles of PNSD and $PM_{2.5}$ concentration around Building A. Error bars denote one standard deviation.

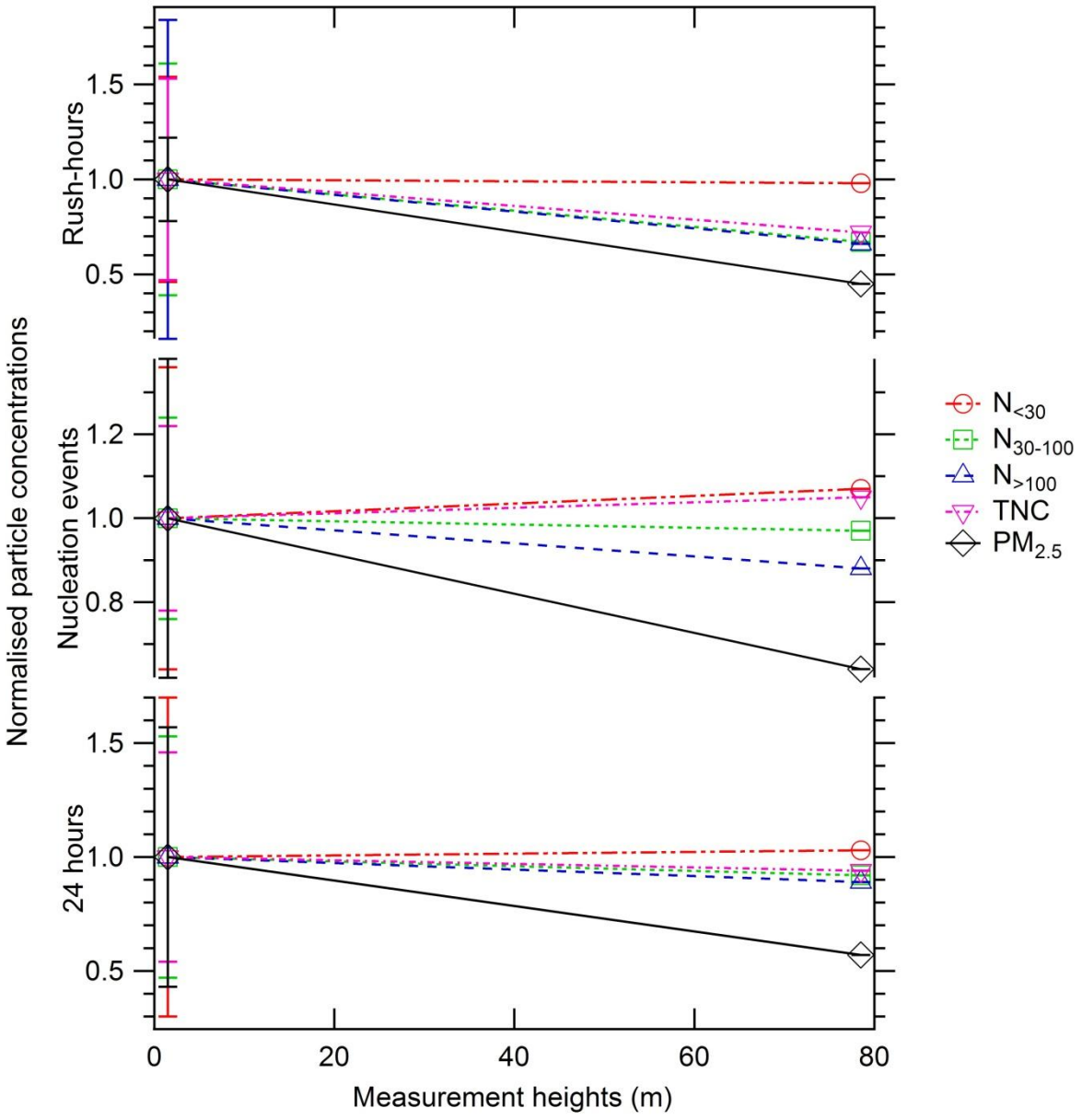

Fig. 11. Vertical profiles of PNSD and $PM_{2.5}$ concentration around Building B. Error bars denote one standard deviation.

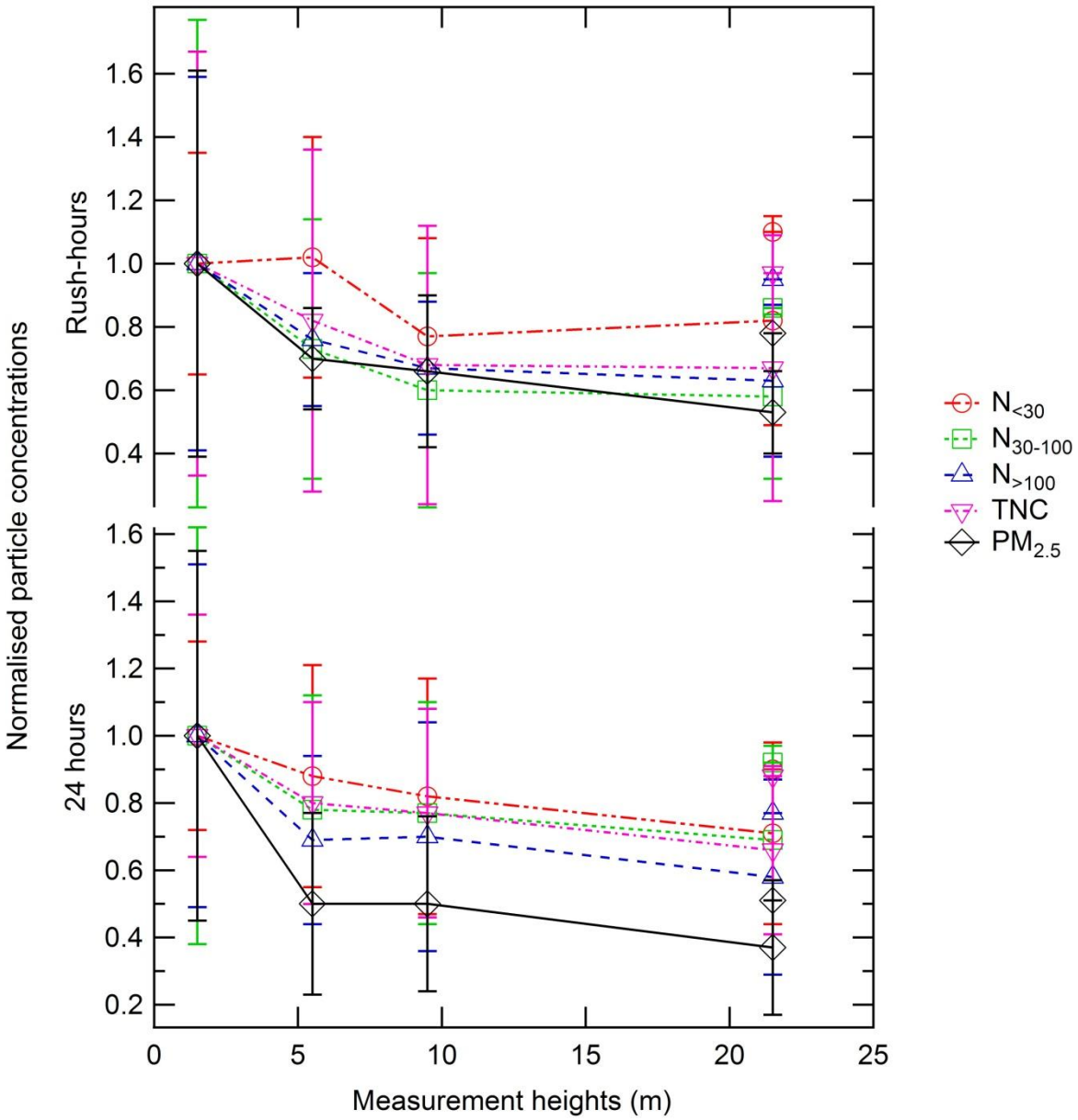

Fig. 12. Vertical profiles of PNSD and $PM_{2.5}$ concentration around Building C. Error bars denote one standard deviation.

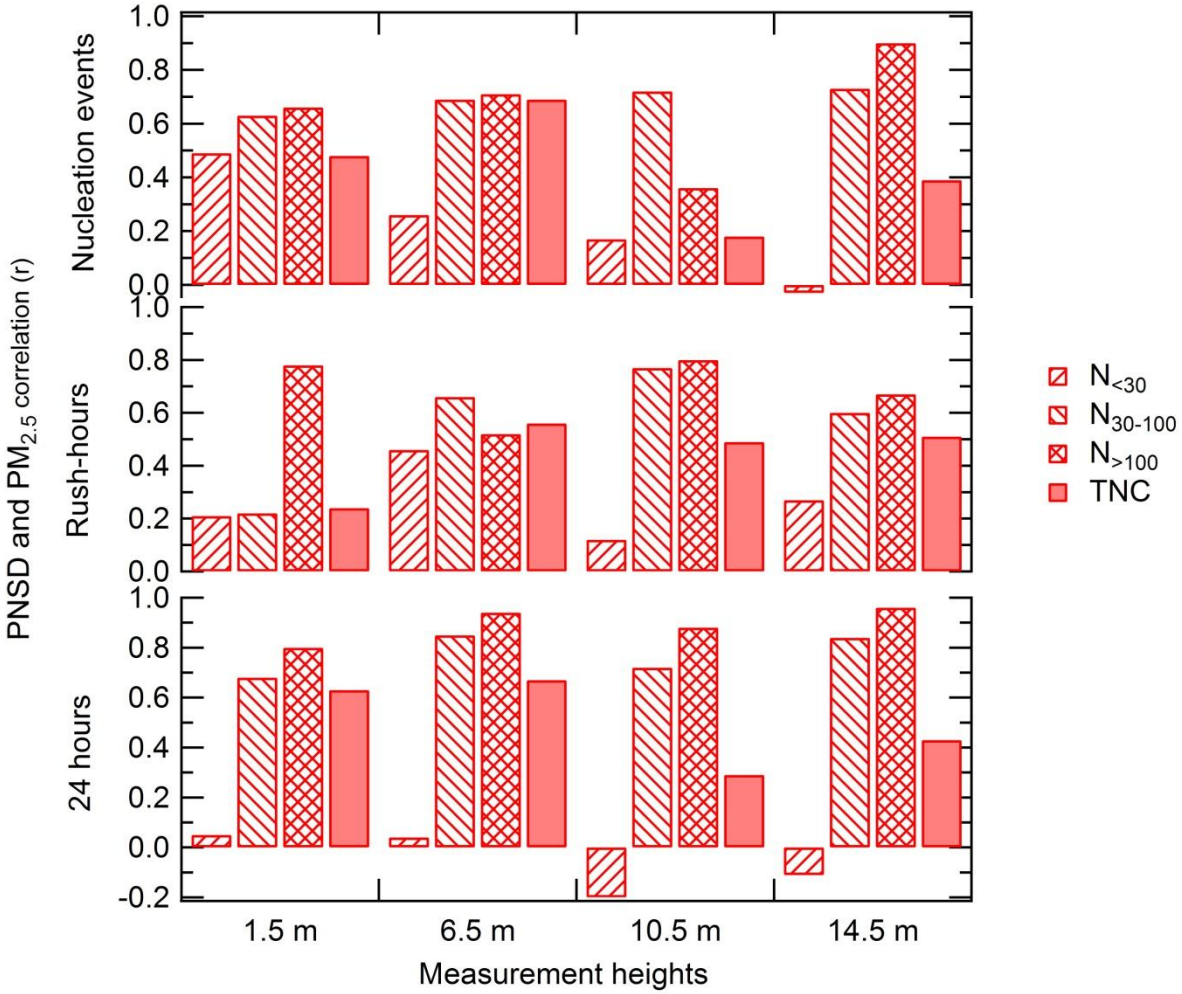

Fig. 13. Relationship between PNSD and $PM_{2.5}$ concentration at different heights for Building A.

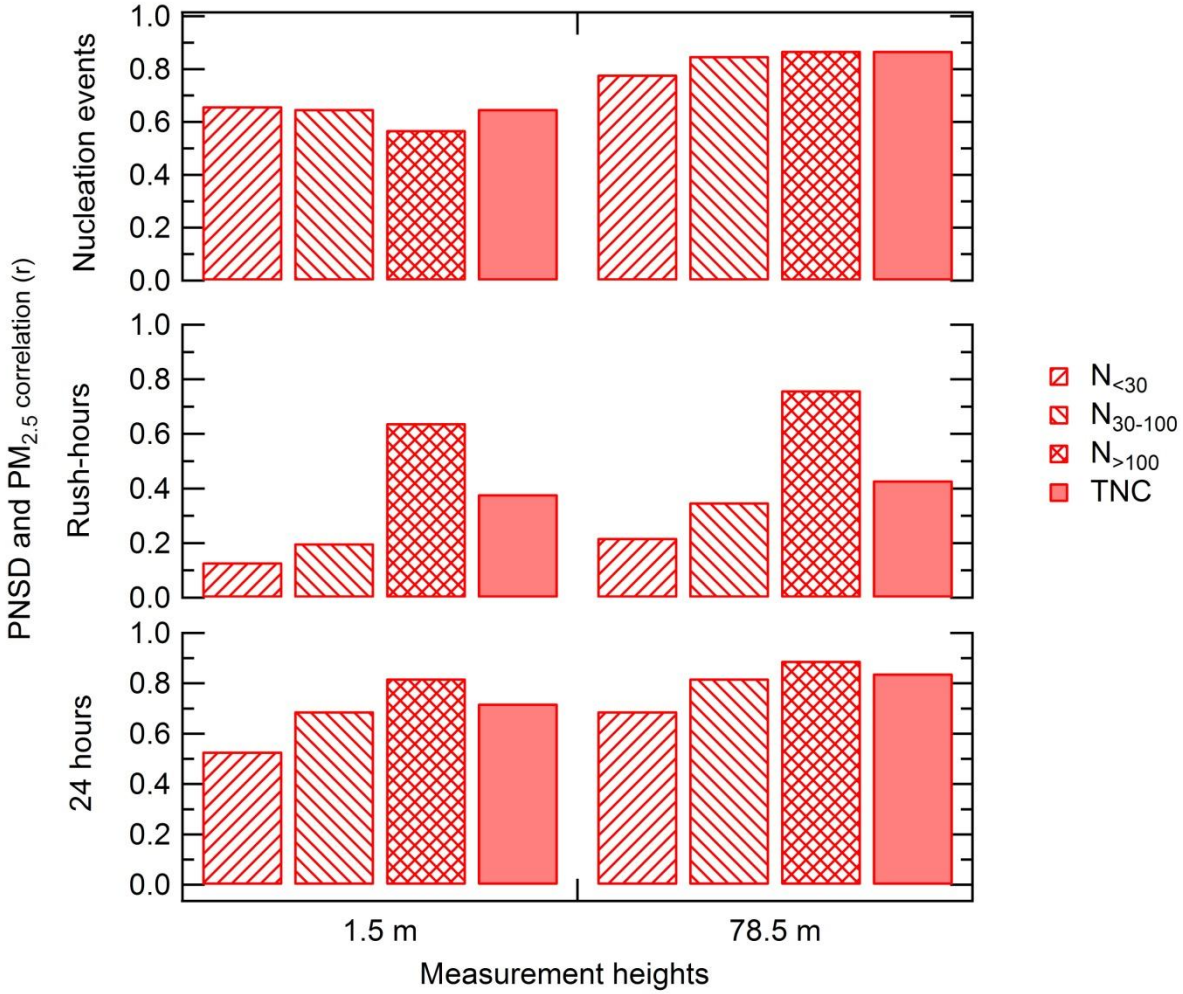

Fig. 14. Relationship between PNSD and $PM_{2.5}$ concentration at different heights for Building B.

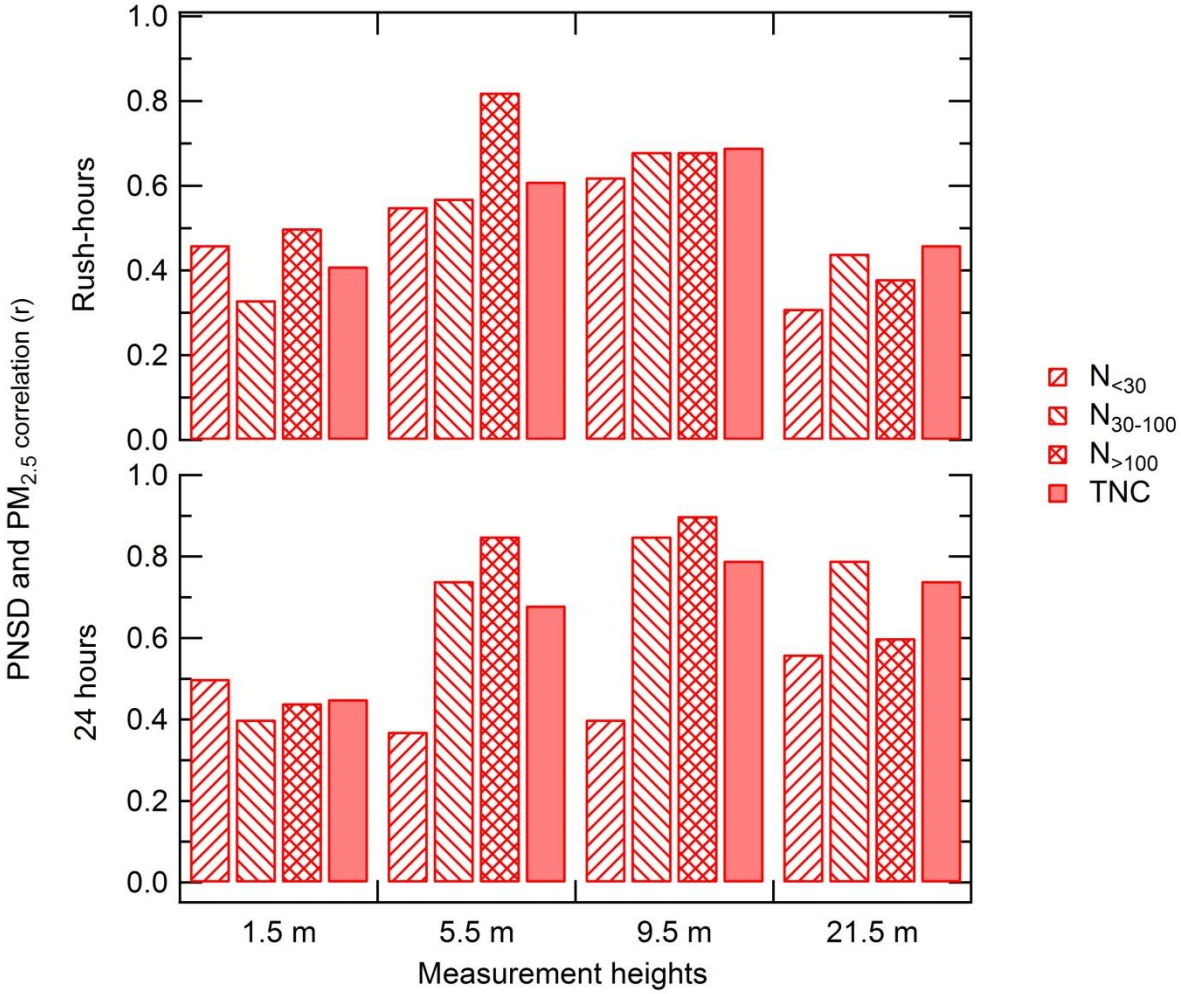

Fig. 15. Relationship between PNSD and $PM_{2.5}$ concentration at different heights for Building C.