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

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Responses to ACPD Referee #1's Comments

Received and published: 05 February 2012

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

General Comments

Comment G1

The authors defined "an event where nucleation mode PN concentrations increased during the day time, but the particles did not grow larger during the event period, as indicated by a near constant GMD value, was considered a nucleation event". The authors also defined the particles with size <30nm as nucleation mode particles. Based on the definitions, sources could directly emit nucleation mode particles in urban areas such as vehicle emissions, aircraft emissions and ship emissions. Nucleation mode particles could also be formed via gas-to-particle conversion (chemical reactions). Although several nucleation events were observed during the sampling weeks for each building, the authors did not provide detailed discussion on the sources of these nucleation events. For instance, could the nucleation events be caused by direct emissions from vehicles as some studies reported PN emitted from vehicles could be smaller than 20 nm, or ships as they exhausted nanoparticles smaller than 10nm in diameter, or something else? Or if the authors believe that these events were due to secondary formation, can they provide more evidence? Through the whole manuscript, they just observed the increase of nucleation mode PN concentrations on some days but did not give any evidence. Solar radiation and temperature were not enough. The question is how do you differentiate primary emissions from secondary formation of nucleation mode particles?

Comment S1.4

. Identification of nucleation event There is a contradictory definition on the event. Suggesting revising (4) the new mode shows signs of growth because immediately another definition of "the new mode does not show signs of growth" was given. More critically, the authors did not clarify whether all events were secondary formation or primary emissions or both. The question is how to differentiate primary from secondary sources.

Response G1 and S1.4

Our research aimed to assess the influence of vehicle emissions and nucleation events in terms of when and how these sources affected the vertical profiles of particle concentration, but not the underlying mechanism of secondary particle formation. Therefore, our intention was to identify the presence, but not the physico-chemical formation of the particles.

To clarify the nucleation event definition, the following paragraph has been added to section 2.5 as an introduction.

"Morawska et al. (2008) has shown that motor vehicle emissions are the major source of air pollution in urban environments. Particles from vehicle emissions are classified as either primary or secondary. The primary particles are generated directly from engines and range in size from 30 – 500 nm. The secondary particles are formed via nucleation in the atmosphere after emissions from the tailpipe and are generally below 30 nm."

And, identification of a nucleation event has been revised as follows:

"In order to identify nucleation events, contour plots of data based on a 24-hour period, from 0:00 – 24:00, were visually analysed. Criteria proposed by Dal Maso et al. (2005) and Hussein et al. (2008) were then applied to identify nucleation events. These criteria are : (i) a distinctly new mode of particles must appear in the size distribution; (ii) the mode starts in size range of < 30 nm; (iii) the mode prevails over a time period of hours; and (iv) the new mode shows signs of growth. In urban environments, nucleation events have been observed both with and without particle growth (Cheung et al., 2011; Gao et al., 2009; Park et al., 2008). Therefore, an event where the nucleation mode particle number concentrations increased during the day, but the particles did not grow larger during the event period, as indicated by a near constant Geometric Mean Diameter (GMD) value, was also considered as a nucleation event. Atmospheric conditions during the events were also recorded to identify the preconditions for nucleation process."

Based on the first and third criteria of the nucleation event definition, it can be seen that it is very difficult to clearly distinguished primary particles in the particle size distribution and they rarely exists for several hours.

A discussion on the sources of the nucleation events at Buildings A, B and C is provided in section 3.2.2 (from para. 5, line 26, page 1624 to para. 2, line 22, page 1627).

It should be noted that local gaseous pollutant data was not collected in our study to assist differentiation of primary from secondary particles as such assessment was not our intention.

Comment G2

It is hard to believe that new particle formation due to chemical reactions can be apparently observed at roadside sites given that emissions from vehicles are fresh and secondary formation needs time to occur. However, primary emissions of nucleation mode particles are possible at roadside sites such as vehicles and nearby sources. In contrast, at ambient sites (sites away from main emission sources) in urban areas secondary particle formation have been widely reported.

Response G2

Previous studies on secondary particles formed from the precursors of vehicle exhaust gases, have been observed near a busy freeway (Charron and Harrison, 2003; Gramotnev and Ristovski, 2004; Harrison et al., 1999; Kittelson et al., 2002; Ntziachristos et al., 2007; Rosenbohm et al., 2005; Sturm et al., 2003; Westerdahl et al., 2005; Zhu et al., 2004) and on-road (Casati et al., 2007; Giechaskiel et al., 2005; Kittelson et al., 2004; Kittelson et al., 2006; Pirjola et al., 2004; Rönkkö et al., 2006; Vogt et al., 2003). Therefore, our observations of the nucleation events at Building A, close to the busway and Building C, close to the freeway are not unusual.

Comment G3

In order to differentiate primary emissions from secondary formation of nucleation mode particles in urban areas, it is obviously not sufficient to measure PN and PM2.5 concentrations. Much more chemicals need to be monitored such as gas-phase primary pollutants CO, NO and SO2, and secondary pollutants such as O3 and SO42-. By comparing the time series of these air pollutants with nucleation mode PN, this problem may be resolved.

Response G3
As we noted in the response G1 and S1.4, O_3, CO, NO and SO_2 were not measured around the building envelopes to distinguish between primary and secondary particles. However, our data has included $PM_{2.5}$ and PN concentrations in different size ranges, and their spectra plots. The nucleation event identification criteria were then used to recognize when vehicle emissions or nucleation events were the dominant source, and how strongly it affected the particle concentration vertical profiles.

Comment G4
Apart from local contributions, regional transport is another important factor affecting the secondary formation. New particles could be formed upwind somewhere else and then be transported to the sites. Though the authors stated that regional transport affected the new particle formation at Building B, it is not convincing that this was caused by a nearby industry zone based on wind direction/speed. Backward trajectory analysis is necessary for the discussion of regional transport impact.

Response G4
In regards to new particle formation at Building B, we have conducted further analysis in accord with the reviewer's comment above. The following text describing this has been added to section 3.2.2., paragraph 3, p. 1626, line 22:

The sentence "... NE of the city. A similar phenomenon ..." has been revised to *" ... NE of the city. Further analysis and comparison of the data measured at this building was conducted along with data collected from a Queensland Department of Environment and Resource Management (DERM) station, which is about 10 km SW of the Brisbane city and 25 km SW of the NE Brisbane industrial zone. The results showed similar trends in PN concentrations between the two locations during the NE winds, but not for other wind directions, during the nucleation days. This implies that emissions from the NE Brisbane industrial zone are those which contribute to the PN concentrations in the Brisbane CBD and surrounding areas. Furthermore, a similar phenomenon ...".*

Specific comments

1. Experimental

Comment S1.1
Sampling locations Figures 1 – 3 did not provide enough information about the sampling sites. It is important to have information on the surrounding environments, especially potential major sources of PN and PM2.5 near the sites. For instance, by checking the Google Earth, I noticed that Brisbane is surrounded by a river. Are there boats on the river? What was the frequency? What fuel was used? Some studies reported that nucleation mode PN emitted from ships was below 10 nm.

Response S1.1
More information describing the measurement sites and potential major pollution sources have been added to section 2.1, paragraph 1, p. 1617, lines 24-26, as follows:

The phrase "We selected ... two million people" has been revised to *"Our research was conducted in the subtropical city of Brisbane, which is the capital city of Queensland, Australia. Further information on the topography and meteorology of this region is described in Cheung et al. (2011). The Brisbane River meanders around the city, and the major air*

pollution sources found in the CBD are inner-city traffic emissions, and aircraft, ship and industrial emissions transported from the lower reaches of the River, located approximately 15-18 km NE of the CBD. We selected three urban office buildings, located close to busy roads with different terrains."

Comment S1.2
sampling My understanding is that the rooftop site was continuously monitored while the lower-level sites were switched in the buildings. Clear information needs to be provided on how the sampling was conducted, such as outdoor air was sampled. Where was the sampling inlet? How far was the sampling inlet from the building walls and so on?

Response S1.2
Further information on the sampling process has been added in section 2.3, paragraph 1, p. 1618, line 26, as follows:

" ... the lower level. Measurement were ..." has been revised to *" ... the lower levels. The air sampled from outdoors (i.e. outside the plant room) was delivered to the instruments via a 1 m long conductive tubing, with an inner diameter of 6 mm. The locations of all outdoor air sampling points were carefully considered to avoid the influence of nearby exhaust air from the HVAC system, if any. A flow splitter was used in cases where several instruments sampled air from the same location. Measurements were ..."*

Comment S1.3
. meteorological data Continuous measurement of meteorological data is critical to understand the new particle formation, if any. By looking at the time series of solar radiation, temperature and winds with pollutants, the possible mechanisms of new particle formation could be explored. And

Comment S2.3
2nd sentence, para 2, p 1622: If the traffic flows on the streets showed corresponding peaks, why not show the daily variations of traffic flow?

Response S1.3 and S2.3
In regards to the above comments, the requested information has been added and the order and title of the following figures has been revised, as follows:

1. Fig. 4 and 5 captions have been revised to:

Fig.5. Daily variation of PNSD and PN size fraction concentrations at Building A.

Fig. S1. Daily variation of PNSD and PN size fraction concentrations at Building B.

Fig. S2. Daily variation of PNSD and PN size fraction concentrations at Building C.

2. Fig. 6 has been revised to:

Fig. 6. PNSD spectra at Building A on a weekday characterised by non- or unclear nucleation events.

3. Fig.7 has been revised to:

Fig. 7. Average particle concentrations and their rooftop to street level ratios at Building A on a weekday characterised by non- or unclear nucleation events.

4. Fig. 8 has been revised to:

Fig. 8. PNSD spectra at Building A on a nucleation event day.

5. Fig. 9 has been revised to:

Fig. 9. Average particle concentrations and their rooftop to street level ratios at Building A, on a nucleation event day.

6. Fig. S1 has been revised to:

Fig. S3. PNSD spectra at Building B on a weekday characterised by the non- or unclear nucleation events.

7. Fig.S2 has been revised to:

Fig. S4. Average particle concentrations and their rooftop to street level ratios at Building B on a weekday characterised by the non- or unclear nucleation events.

8. Fig. S3 has been revised to:

Fig. S5. PNSD spectra at Building C on a weekday characterised by the non- or unclear nucleation events.

9. Fig.S4 has been revised to:

Fig. S6. Average particle concentrations and their rooftop to street level ratios at Building C on a weekday characterised by the non- or unclear nucleation events.

10. Fig. S5 has been revised to:

Fig. S7. PNSD spectra at Building B on a nucleation event day.

11. Fig. S6 has been revised to:

Fig. 8. Average particle concentrations and their rooftop to street level ratios at Building B on a nucleation event day.

12. Fig. S7 has been revised to:

Fig. S9. PNSD spectra at Building C on a nucleation event day.

13. Fig. S8 has been revised to:

Fig. S10. Average particle concentrations and their rooftop to street level ratios at Building C on a nucleation event day.

The traffic flow rates on the streets close to the sampling sites at Buildings A, B, and C have been added to the new figs. 5, S1, and S2, respectively. Wind direction and speed, solar

radiation, and temperature and humidity have been added to the new figs. 6, 8, S3, S5, S7, and S9. These figures are illustrated in appendixes.

2. Results

Comment S2.1
It is not common to say "particle size distribution concentrations". It may be said either "particle size distribution and concentration" or "nucleation mode, Aitken mode and accumulation mode particle concentration"

Response S2.1
The phrase 'Particle number size distribution concentrations' has been revised to '*Particle number size distribution - PNSD*'.

Comment S2.2
1st sentence, para 2, page 1622: It is not obvious for Building B.

Response S2.2
The 1st sentence of paragraph 2, page 1622, lines 7-8 has been revised to *"Daily mean variations of PN size fraction concentrations increased in the early morning and late afternoon at Buildings A and C."*

Comment S2.4
3rd sentence, para 2, p1622: be careful when the statement of ": : :suggests the occurrence of new particle formation" was given as it could be either primary or secondary emissions.

Response S2.4
Primary particles generated from the engine often range in size from 30 to 500 nm, while secondary particles formed outside, are generally in the range below 30 nm (Morawska et al., 2008). Only the PN concentration in the size range of < 30 nm, increased when the other size ranges decreased. In addition, the traffic flow rates on the streets close to the sampling sites decreased during the midday period, which could further suggest the occurrence of newly formed particles. To clarify this, the following has been added to the section 3.1., paragraph. 4, p. 1622, lines 11 – 13:

The sentence "In contrast, ... of new particle formation." has been revised to *"In contrast, $N_{<30}$ concentration increased at noon, while other particle size ranges remained constant or decreased at both the rooftop and street levels of all three buildings. In addition, the traffic flow rates decreased around midday. This could suggest the occurrence of new particle formation during this period."*

Comment S2.5
last para, page 1622: how many weekdays were characterised by absent or unclear nucleation events for each site? How do you define unclear nucleation events?

Response S2.5
We measured 26, 17, and 23 days at Building A, B and C, respectively (refer to section 2.3). We identified 7, 9, and 3 nucleation days at Building A, B and C, respectively (refer to section 3.2.2). So there were 19, 8, and 20 non- or unclear nucleation event days at Building A, B, and C, respectively.

To clarify this, the first sentence of paragraph 1, in section 3.2.1 ‘Based on ... each building.’ has been revised to *‘The days that did not meet at least one of the criteria for the nucleation event definition were defined as a non- or unclear nucleation event day. Based on this, there were 19, 8, and 20 days that were classified as non- or unclear nucleation event at Building A, B, and C, respectively. Weekdays characterised by non- or unclear nucleation events were selected to assess the influence of vehicle emissions on the PN and $PM_{2.5}$ concentrations at the rooftop and street levels of each building.”*

Comment S2.6
3.2.2 Influence of new particle formation on : : :: : :. From figs 8, 9 S5-8, it is hard to believe new particle formation occurred because the PN concentration from 8.5 – 15 nm was almost nil, and no other evidence i.e. O3, CO, NO and SO2 measurement data was given. Also, unlike described in the manuscript, the accumulation-mode PN appeared to have good correlation with nucleation-mode PN in these figs, meaning primary source emissions with different size of particles.

Response S2.6
Our data presented in figs. 8, 9, S5-8 (the new figs. 8,9, S7-10) show that the PN in both size ranges of < 30 nm and 30 – 100 nm, and $PM_{2.5}$ concentrations increased during the rush hour periods. However, only $N_{< 30}$ concentration increased, while both $N_{30\text{-}100}$ and $PM_{2.5}$ concentrations decreased during the midday period. Simultaneously, the spectra of particle size distribution clearly show the occurrence of nucleation events during these times.

Comment S2.7
1st and 2nd paras, p1625: it could be true that the N<30 and N<30/N30-300 at the rooftop were higher than those at ground levels. However, the reasons may not be right as the chemistry at rooftop may be totally different from that at roadside. At roadside, chemical reactions for new particle formation in the atmosphere would be very limited due to highly fresh emissions and constrained oxidant concentrations i.e. O3 and OH. Hence, most likely the N<30 was related to direct emission at ground level while the air mass at rooftop could have chemical reactions to form new particles plus vertical diffusion of primary nucleation mode particles from the streets. Also, it should be careful that the primary pollutants involving in the new particle formation at rooftop could originate from urban vehicle emissions (not the immediate ones near the sampling sites) and/or from regional transport. To thoroughly understand the mechanisms, solar radiation and wind direction are certainly not sufficient.

Response S2.7
As discussed in the response G2 above, new particle formation from vehicle emissions at the roadside, are often observed (Morawska et al., 2008). As we noted, the major pollution sources surrounding Buildings A and C were the high traffic flow busway and freeway, respectively. The occurrences of nucleation events, due to local vehicle emissions at these buildings were therefore not unusual. The formation process found at the street and rooftop levels of these buildings was expected to be dependant mainly on local conditions, such as high condensable gas concentrations and solar radiation intensity, together with low pre-existing particle concentrations. Conditions for locally new particle formation were also reported in previous research by Vakeva et al. (1999) and Kumar et al. (2009). Meanwhile, at Building B the newly formed particles were transported from a nearby industrial zone and therefore, new particle production was not the result of local sources but was strongly influenced by wind speed, wind direction and the origin of incoming air masses (refer to section 3.2.2 for more detail discussions).

Comment S2.8
1st para, page 1626: could the ships nearby be one of the causes?

Response S2.8
In regards to the emission sources around Building C, as denoted in the site description, the Brisbane River meanders past the building. However, emissions from the river (with about 250 ferries per day) are minimal compared to vehicle emissions from the freeway (110,000 vehicles per day).

Comment S2.9
2nd para, p1626: the statement on statistical difference in solar radiation was wrong. P=0.36 means there was no difference between the two sites. Similarly, throughout the entire manuscript, there were many statistical descriptions. But when looked at the mean _ S.D. and p values, sometimes we don't know which one we should trust. For instance, 1.15_0.35 vs. 1.88_1.19, large deviations suggest these two values did not have statistical difference. However, the test for this pair was p<0.001, suggesting significant difference. Generally mean _ 95% confidence interval would avoid this confusion.

Response S2.9
The statement on statistical difference in solar radiation has been revised, and Mean ± SD have been modified to Mean ± 95% CI.

In section 3.2.2, paragraph 7, p. 1626, lines 9-16, the sentence "Based on ... , respectively)." has been revised to *"Based on $N_{<30}$ and $N_{<30}/N_{30-300}$ at rooftop and street levels, we also concluded that the intensity of new particle formation at Building B on 16 January 2010, was clearly stronger than that at Buildings A and C, although the mean solar radiation intensity ($W\ m^{-2}$) (Mean ± 95% CI) during the nucleation event at Building B was not significantly different compared to Building A (664.3 ± 20.7 vs. 689.4 ± 22.4, p = 0.36). At the same time, ratios between rooftop and street level values for $N_{<30}$ and $N_{<30}/N_{30-300}$ were significantly lower at Building B compared to those at Building A (1.15 ± 0.09 vs. 1.88 ± 0.27, p<0.01; 1.20 ± 0.14 vs. 1.84 ± 0.30, p < 0.01, respectively)."*

Comment S2.10
last para, page 1626 and 1st para, page 1627: From Table 3, there was a significant difference in PN between the rooftop and street level at Building B (p = 0.01). However, here it said no difference. Is this for 16 Jan 2010 one day? If so, give PN concentrations at both sites and p-value.

Response S2.10
In section 3.2.2, paragraph 8, p. 1627, lines 1-2, the sentence "there was ... at Building B." has been revised to *"the difference in PN concentrations (cm^{-3}) between the rooftop and street levels at Building B (16,900 ± 1,490 vs. 15,650 ± 1,470; p < 0.05) was significant, but not to the same extent observed at Buildings A (8,160 ± 1,020 vs. 4,570 ± 280; p < 0.01) and C (5,340 ± 450 vs. 3.310 ± 270; p < 0.01)."*

Comment S2.11
3.3 vertical profiles of particle concentrations For Building B, caution should be taken for vertical profile discussion as only two points were measured. We don't know what could happen between these two points.

Response S2.11
Please refer to the response G8 and S9 for the Referee 2 for revisions related to the vertical profiles at Building B.

Comment S2.12
Table 2: it would be helpful if the number of days can be listed for each building when vehicle emissions dominated. Mean _ SD should be replaced by mean _ 95% Confidence Interval. It is strange that p-value representation was not consistent. For instance, sometimes p<0.001 but sometimes p = 0.002, 0.006- both should be p<0.01. For p=0.015, it should be p<0.05.

Response S2.12
The mean ± SD and p_values in Table 2 have been revised to:

Table 2. Average particle concentrations at the rooftop and the street levels of Buildings A, B and C during rush-hour periods.

Site	Level	PN (Mean ± 95% CI) × 10^3 (cm^{-3})			$PM_{2.5}$ (Mean ± 95% CI) (μg m^{-3})		
		Morning	Afternoon	p	Morning	Afternoon	p
Building A	Rooftop	18.73 ± 1.21	9.99 ± 0.73	< 0.01	42.90 ± 1.74	10.10 ± 0.62	< 0.01
	Street	14.51 ± 0.85	7.56 ± 0.43	< 0.01	78.50 ± 3.69	11.80 ± 0.86	< 0.01
	p	< 0.01	< 0.01		< 0.01	< 0.01	
Building B	Rooftop	5.01 ± 0.37	5.82 ± 0.64	< 0.05	8.51 ± 0.48	9.59 ± 0.27	< 0.01
	Street	6.04 ± 0.65	7.21 ± 0.69	< 0.05	19.64 ± 1.14	22.02 ± 1.22	< 0.01
	p	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	
Building C	Rooftop	18.64 ± 1.21	8.56 ± 0.65	< 0.01	19.00 ± 0.51	8.00 ± 0.67	< 0.01
	Street	12.48 ± 1.70	8.12 ± 0.52	< 0.01	17.70 ± 0.79	8.20 ± 0.56	< 0.01
	p	< 0.01	0.06		< 0.05	0.45	

Comment S2.13
Table 13: same problems as Table 2.

Response S2.13
The mean ± SD and p_values in Table 3 have been revised to:

Table 3. Average particle concentrations during the nucleation event days.

Site	Level	$N_{<30}$ (cm^{-3}) (Mean ± 95% CI) × 10^3	$N_{<30}/N_{30-300}$ (Mean ± 95% CI)	$PM_{2.5}$ (μg m^{-3}) (Mean ± 95% CI)
Building A	Rooftop	8.16 ± 1.02	1.76 ± 0.33	11.34 ± 1.11
	Street	4.57 ± 0.28	1.01 ± 0.08	19.74 ± 3.50
	p	< 0.01	< 0.01	< 0.01
Building B	Rooftop	16.90 ± 1.49	4.54 ± 0.52	4.0 ± 0.08
	Street	15.65 ± 1.47	3.92 ± 0.34	7.5 ± 0.65
	p	< 0.05	< 0.01	< 0.01
Building C	Rooftop	5.34 ± 0.45	2.23 ± 0.32	1.67 ± 0.18
	Street	3.31 ± 0.27	1.91 ± 0.24	2.01 ± 0.14
	p	< 0.01	<0.01	< 0.01

References

Casati, R., Scheer, V., Vogt, R., and Benter, T.: Measurement of nucleation and soot mode particle emission from a diesel passenger car in real world and laboratory in situ dilution, Atmospheric Environment, 41, 2125-2135, 2007.

Charron, A., and Harrison, R. M.: Primary particle formation from vehicle emissions during exhaust dilution in the roadside atmosphere, Atmospheric Environment, 37, 4109-4119, 2003.

Cheung, H. C., Morawska, L., and Ristovski, Z. D.: Observation of new particle formation in subtropical urban environment, Atmospheric Chemistry and Physics, 11, 3823-3833, 10.5194/acp-11-3823-2011, 2011.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen, K. E. J.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiala, Finland, Boreal Environment Research, 10, 323-336, 2005.

Giechaskiel, B., Ntziachristos, L., Samaras, Z., Scheer, V., Casati, R., and Vogt, R.: Formation potential of vehicle exhaust nucleation mode particles on-road and in the laboratory, Atmospheric Environment, 39, 3191-3198, 2005.

Gramotnev, G., and Ristovski, Z.: Experimental investigation of ultra-fine particle size distribution near a busy road, Atmospheric Environment, 38, 1767-1776, 2004.

Harrison, R. M., Jones, M., and Collins, G.: Measurements of the physical properties of particles in the urban atmosphere, Atmospheric Environment, 33, 309-321, 1999.

Hussein, T., Martikainen, J., Junninen, H., Sogacheva, L., Wagner, R., Dal Maso, M., Riipinen, I., Aalto, P. P., and Kulmala, M.: Observation of regional new particle formation in the urban atmosphere, Tellus B, 60, 509-521, 10.1111/j.1600-0889.2008.00365.x, 2008.

Kittelson, D. B., Watts, W. F., and Johnson, J.: Diesel aerosol sampling methodology - CRC E-43: final report, University of Minnesota, Report for the Coordinating Research Council, 2002.

Kittelson, D. B., Watts, W. F., and Johnson, J. P.: Nanoparticle emissions on Minnesota highways, Atmospheric Environment, 38, 9-19, 2004.

Kittelson, D. B., Watts, W. F., Johnson, J. P., Schauer, J. J., and Lawson, D. R.: On-road and laboratory evaluation of combustion aerosols—Part 2:: Summary of spark ignition engine results, Journal of Aerosol Science, 37, 931-949, 2006.

Kumar, P., Fennell, P. S., Hayhurst, A. N., and Britter, R. E.: Street Versus Rooftop Level Concentrations of Fine Particles in a Cambridge Street Canyon, Boundary-Layer Meteorology, 131, 3-18, 10.1007/s10546-008-9300-3, 2009.

Morawska, L., Ristovski, Z., Jayaratne, E. R., Keogh, D. U., and Ling, X.: Ambient nano and ultrafine particles from motor vehicle emissions: Characteristics, ambient processing and implications on human exposure, Atmospheric Environment, 42, 8113-8138, 2008.

Ntziachristos, L., Ning, Z., Geller, M. D., and Sioutas, C.: Particle Concentration and Characteristics near a Major Freeway with Heavy-Duty Diesel Traffic, Environmental Science & Technology, 41, 2223-2230, 10.1021/es062590s, 2007.

Pirjola, L., Parviainen, H., Hussein, T., Valli, A., Hämeri, K., Aaalto, P., Virtanen, A., Keskinen, J., Pakkanen, T. A., Mäkelä, T., and Hillamo, R. E.: "Sniffer"—a novel tool for chasing vehicles and measuring traffic pollutants, Atmospheric Environment, 38, 3625-3635, 2004.

Rönkkö, T., Virtanen, A., Vaaraslahti, K., Keskinen, J., Pirjola, L., and Lappi, M.: Effect of dilution conditions and driving parameters on nucleation mode particles in diesel exhaust: Laboratory and on-road study, Atmospheric Environment, 40, 2893-2901, 2006.

Rosenbohm, E., Vogt, R., Scheer, V., Nielsen, O. J., Dreiseidler, A., Baumbach, G., Imhof, D., Baltensperger, U., Fuchs, J., and Jaeschke, W.: Particulate size distributions and mass measured at a motorway during the BAB II campaign, Atmospheric Environment, 39, 5696-5709, 2005.

Sturm, P. J., Baltensperger, U., Bacher, M., Lechner, B., Hausberger, S., Heiden, B., Imhof, D., Weingartner, E., Prevot, A. S. H., Kurtenbach, R., and Wiesen, P.: Roadside measurements of particulate matter size distribution, Atmospheric Environment, 37, 5273-5281, 2003.

Väkevä, M., Hämeri, K., Kulmala, M., Lahdes, R., Ruuskanen, J., and Laitinen, T.: Street level versus rooftop concentrations of submicron aerosol particles and gaseous pollutants in an urban street canyon, Atmospheric Environment, 33, 1385-1397, 1999.

Vogt, R., Scheer, V., Casati, R., and Benter, T.: On-Road Measurement of Particle Emission in the Exhaust Plume of a Diesel Passenger Car, Environmental Science & Technology, 37, 4070-4076, 10.1021/es0300315, 2003.

Westerdahl, D., Fruin, S., Sax, T., Fine, P. M., and Sioutas, C.: Mobile platform measurements of ultrafine particles and associated pollutant concentrations on freeways and residential streets in Los Angeles, Atmospheric Environment, 39, 3597-3610, 2005.

Zhu, Y. F., Hinds, W. C., Shen, S., and Sioutas, C.: Seasonal trends of concentration and size distribution of ultrafine particles near major highways in Los Angeles, Aerosol Sci Technol, 38, 5-13, 2004.

Appendices

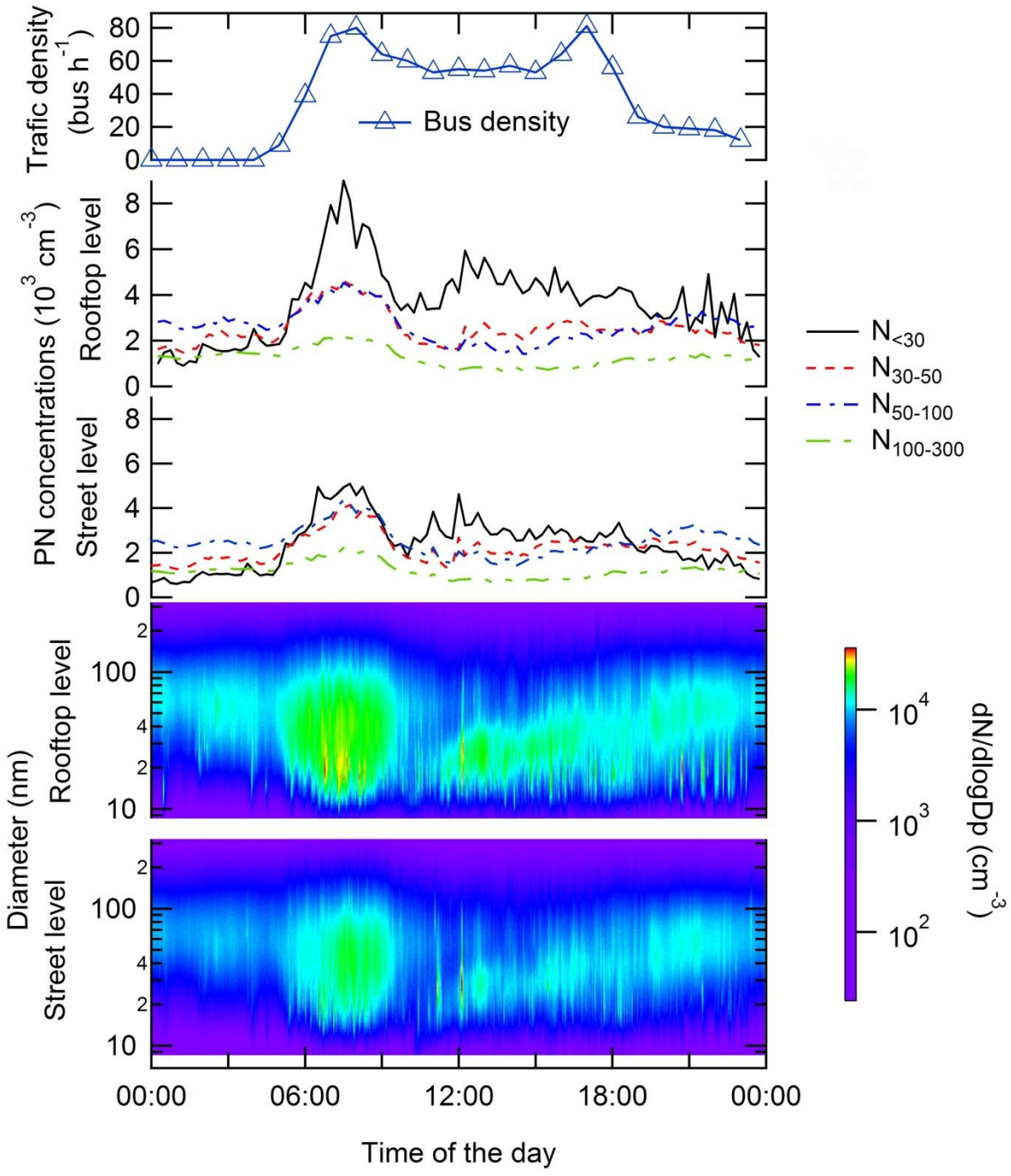

Fig. 5. Daily variation in PNSD and PN size fraction concentrations at Building A.

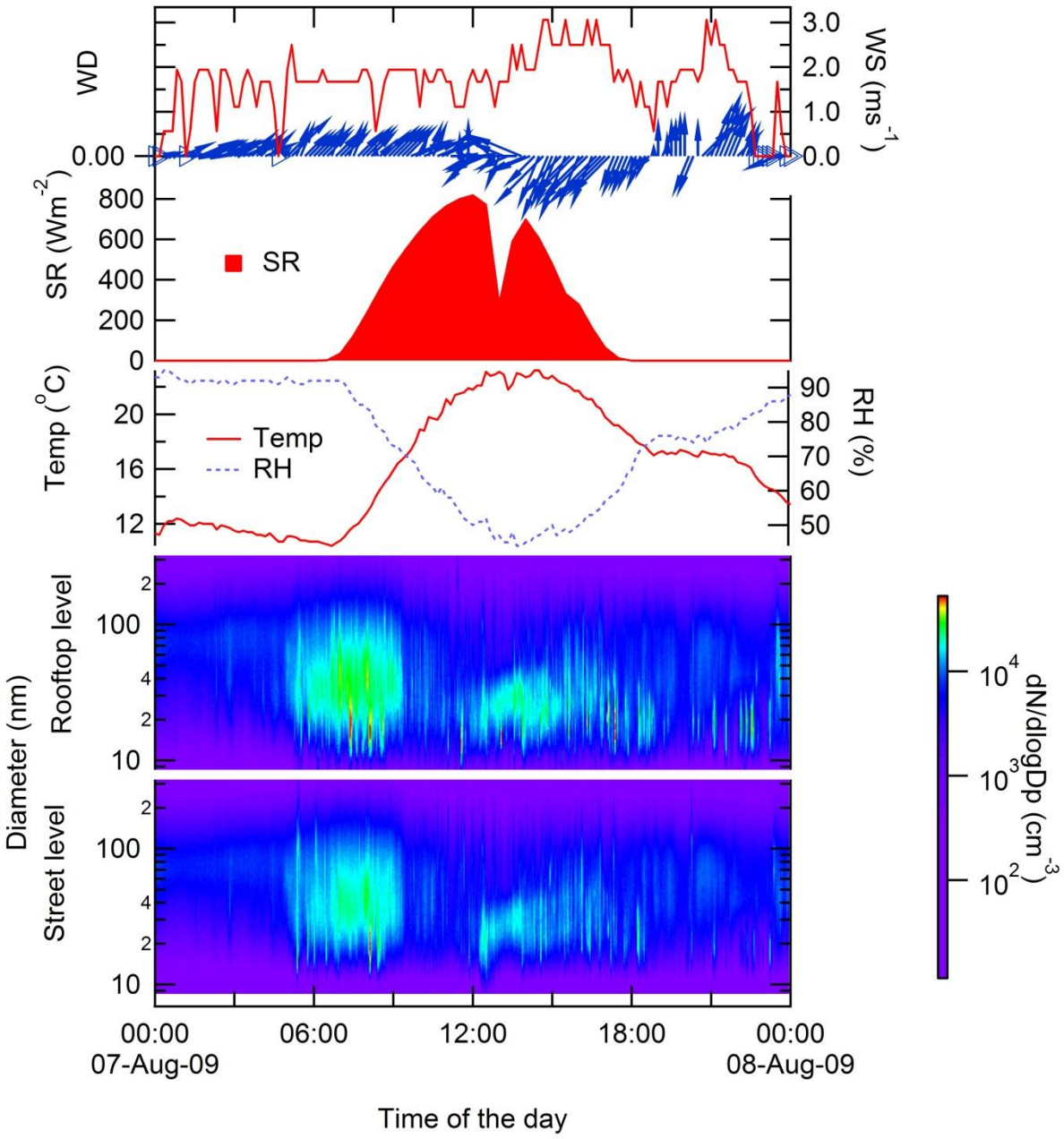

Fig. 6. PNSD spectra at Building A on a week day characterised by the non- or unclear nucleation events.

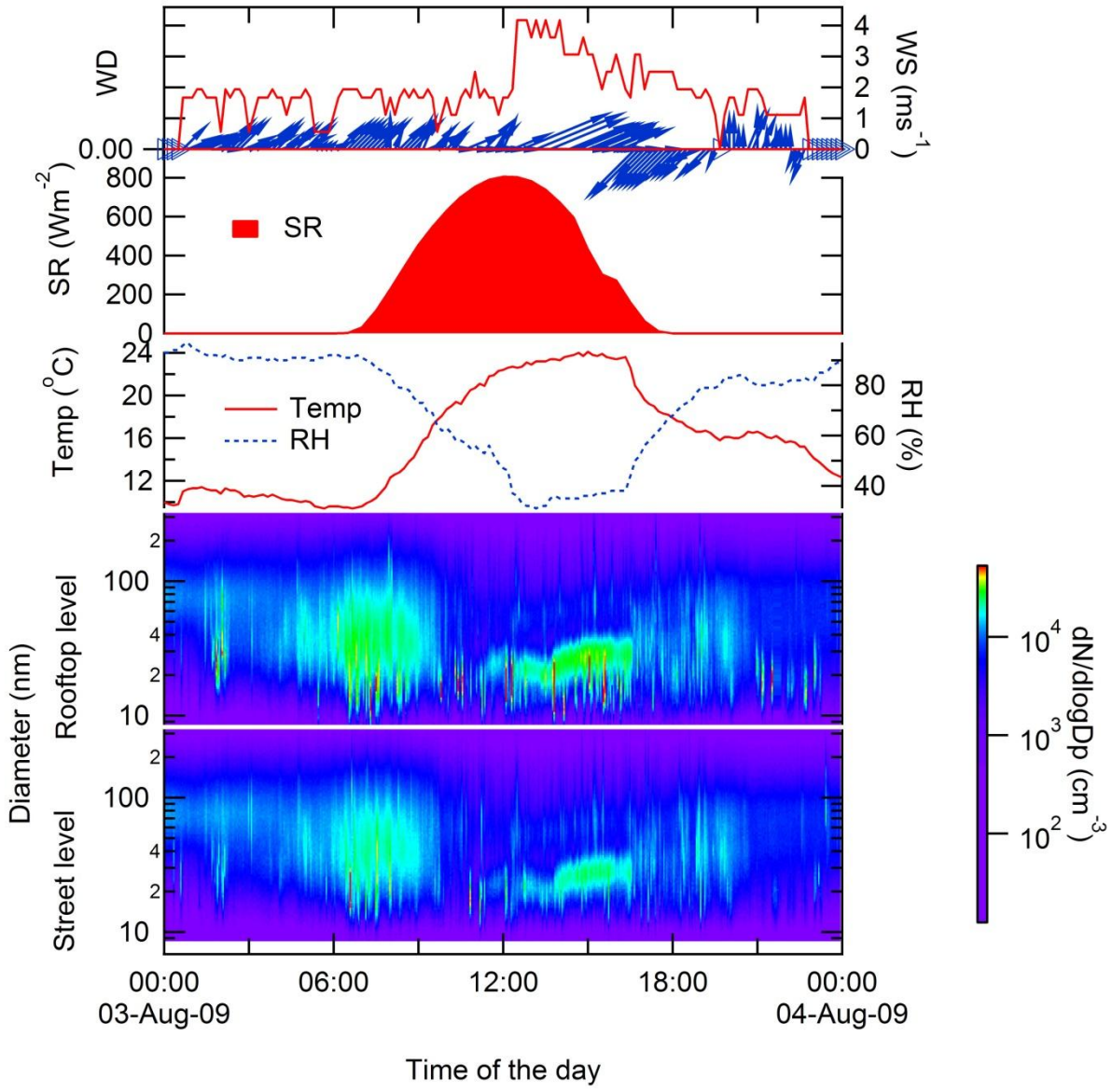

Fig. 8. PNSD spectra at Building A on a nucleation event day.

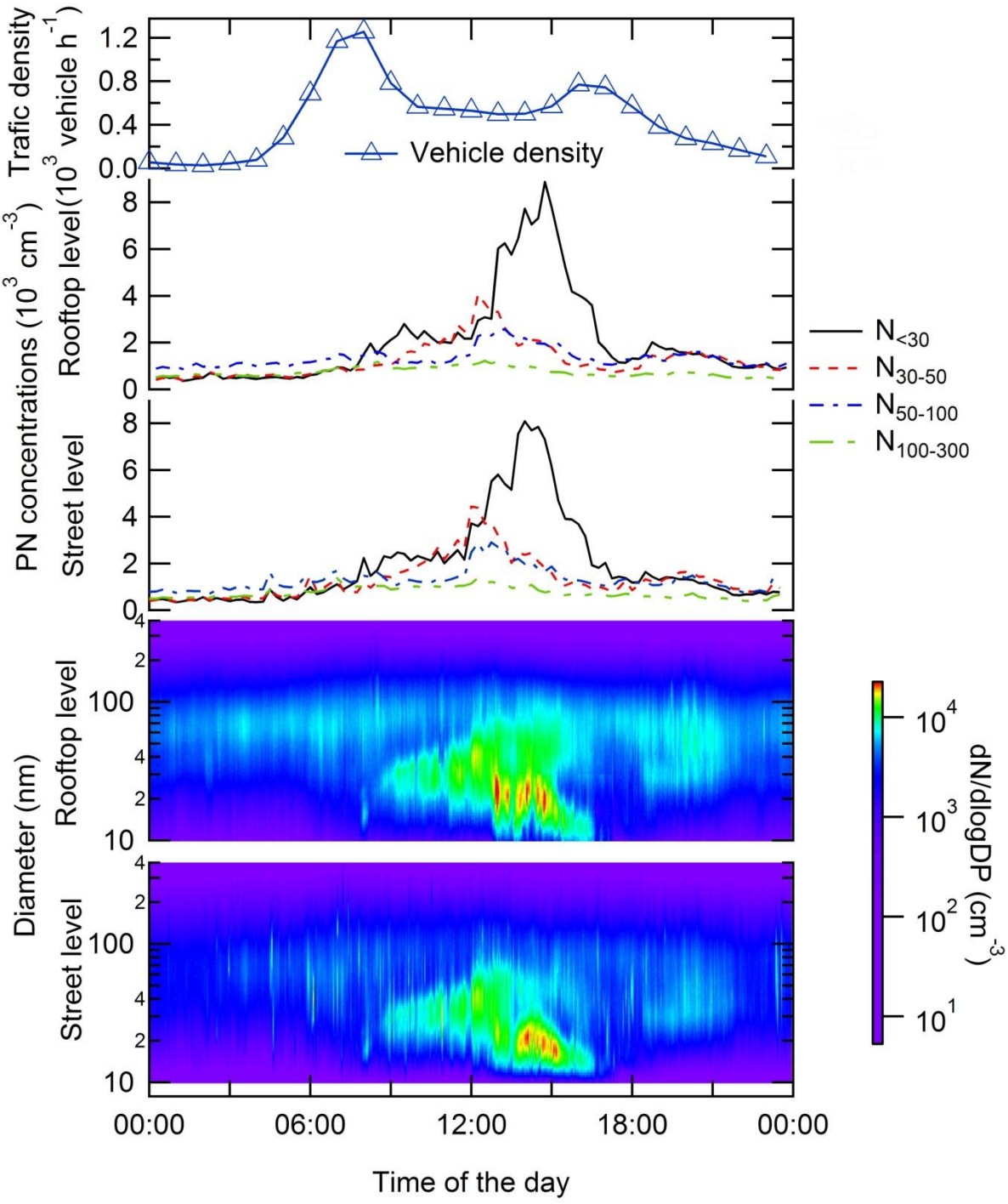

Fig. S1. Daily variation of PNSD and PN size fraction concentrations at Building B.

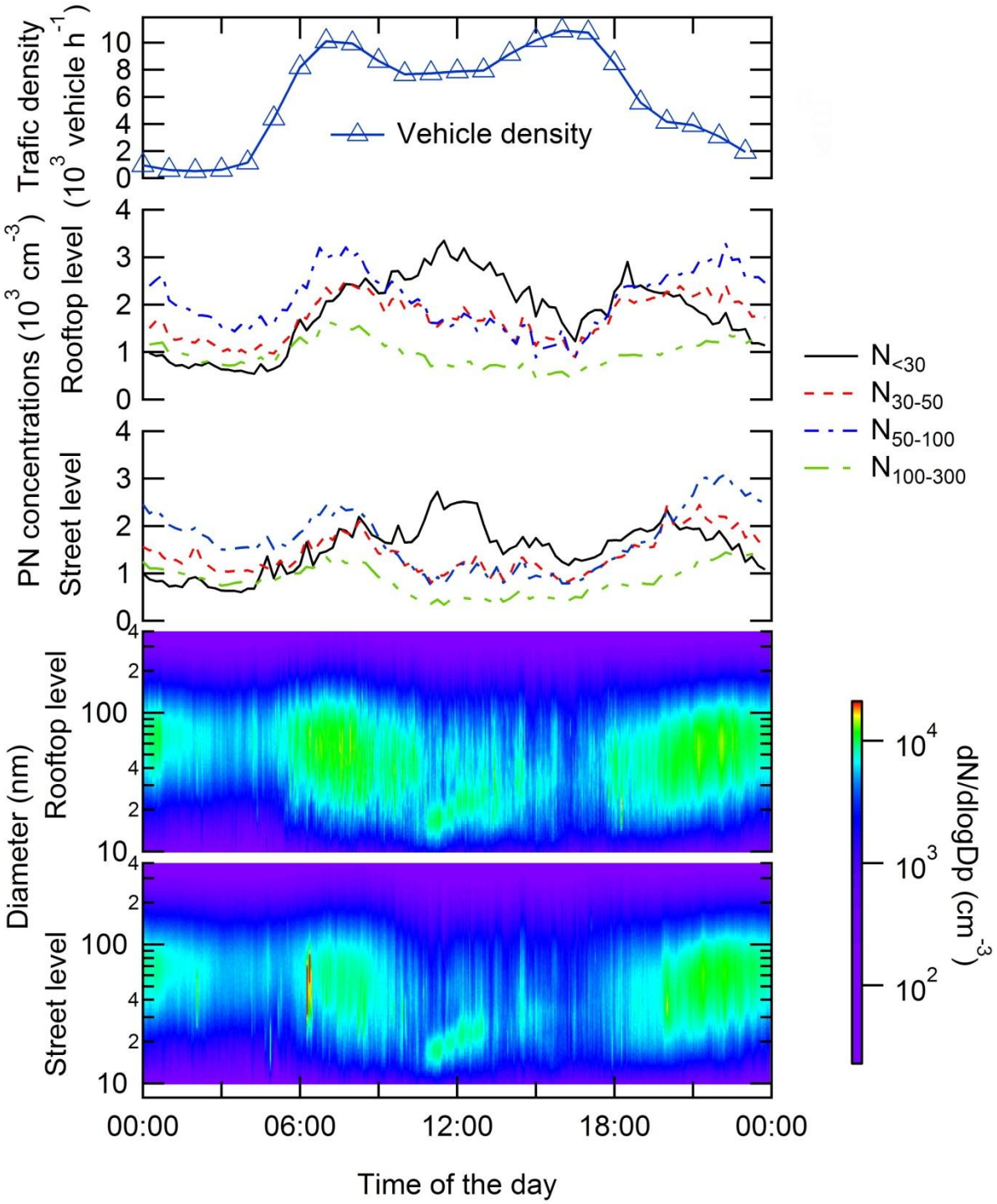

Fig. S2. Daily variation of PNSD and PN size fraction concentrations at Building C.

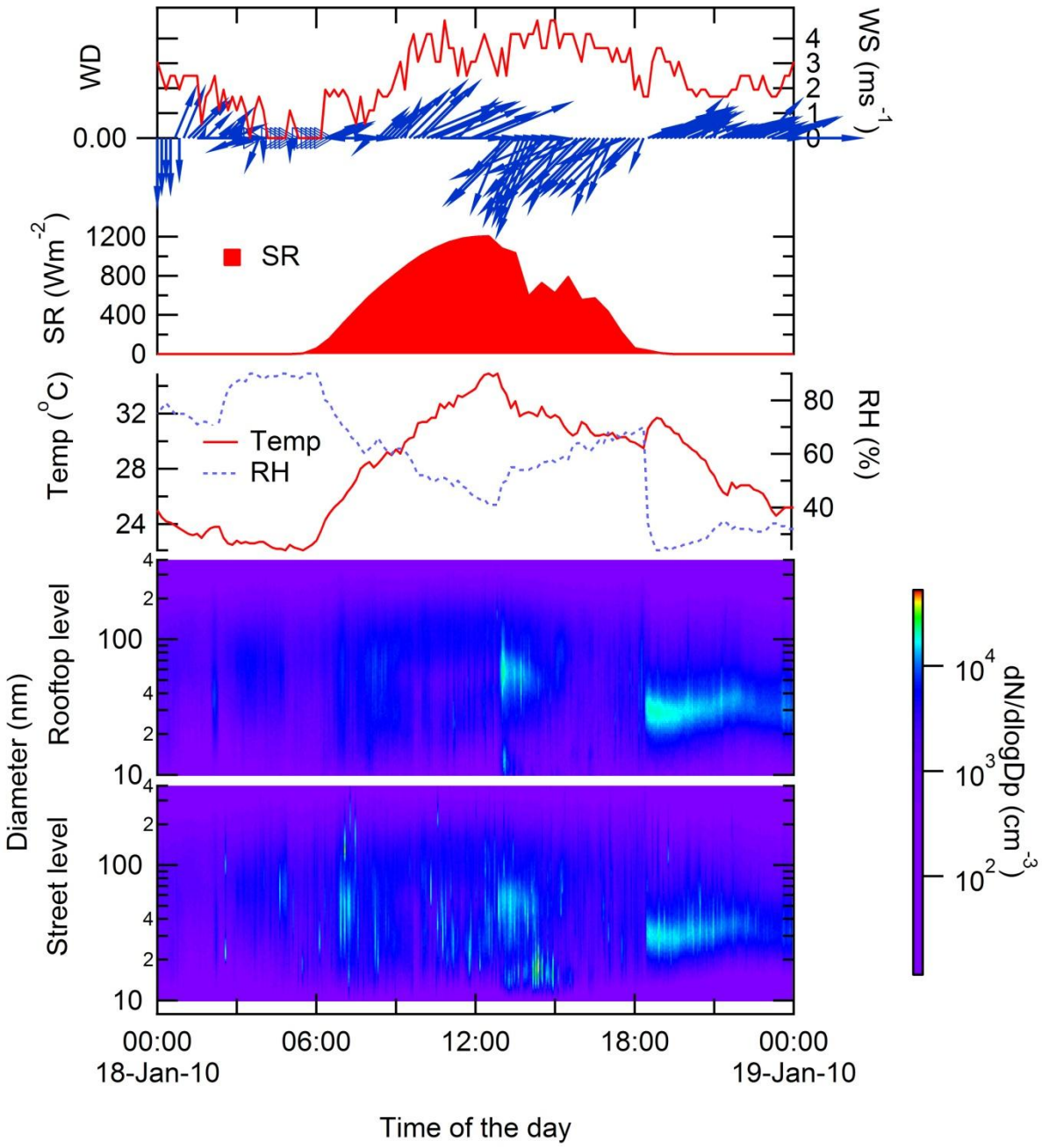

Fig. S3. PNSD spectra at Building B on a week day characterised by the non- or unclear nucleation events.

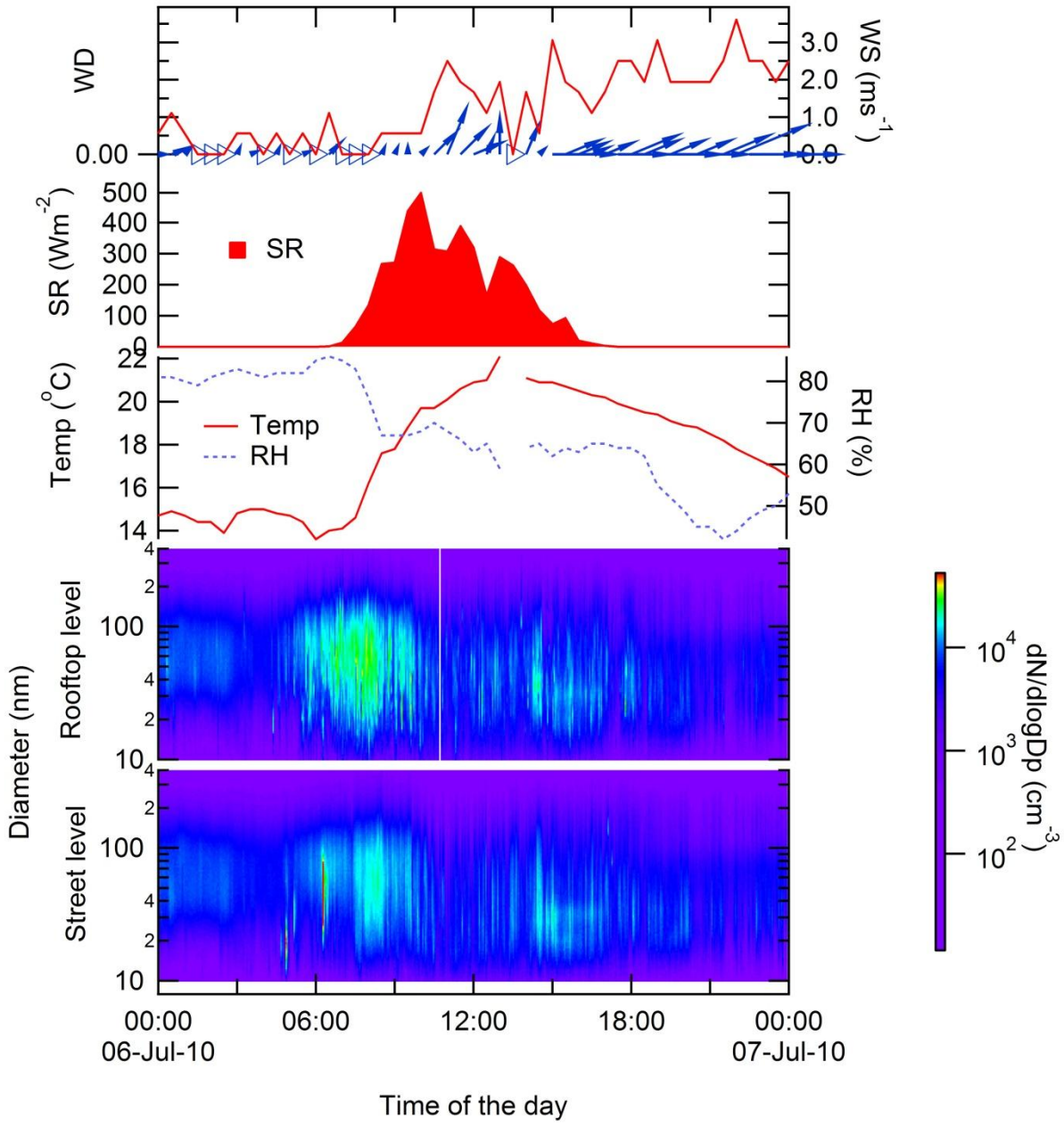

Fig. S5. PNSD spectra at Building C on a weekday characterised by the non- or unclear nucleation events.

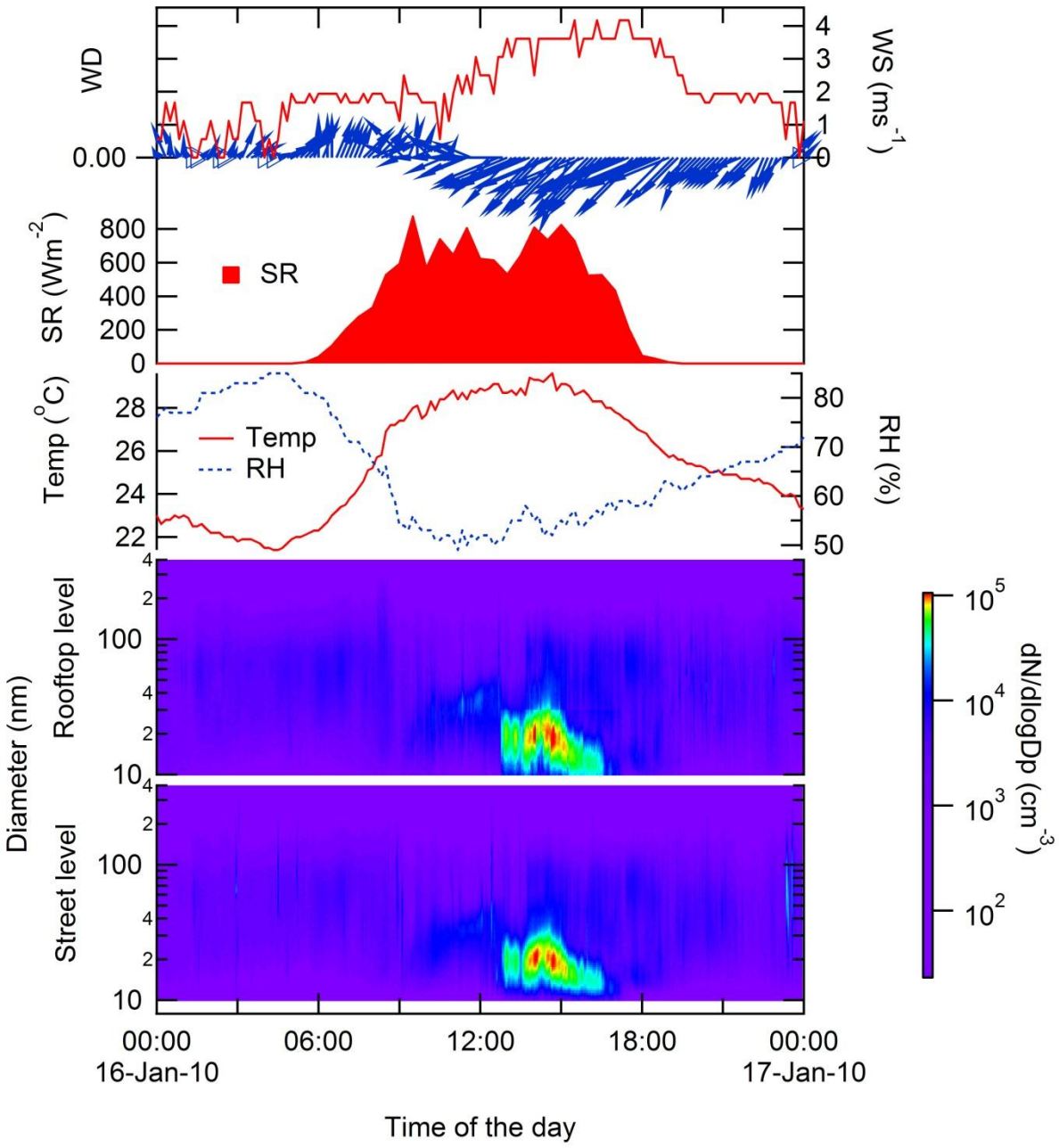

Fig. S7. PNSD spectra at Building B on a nucleation event day.

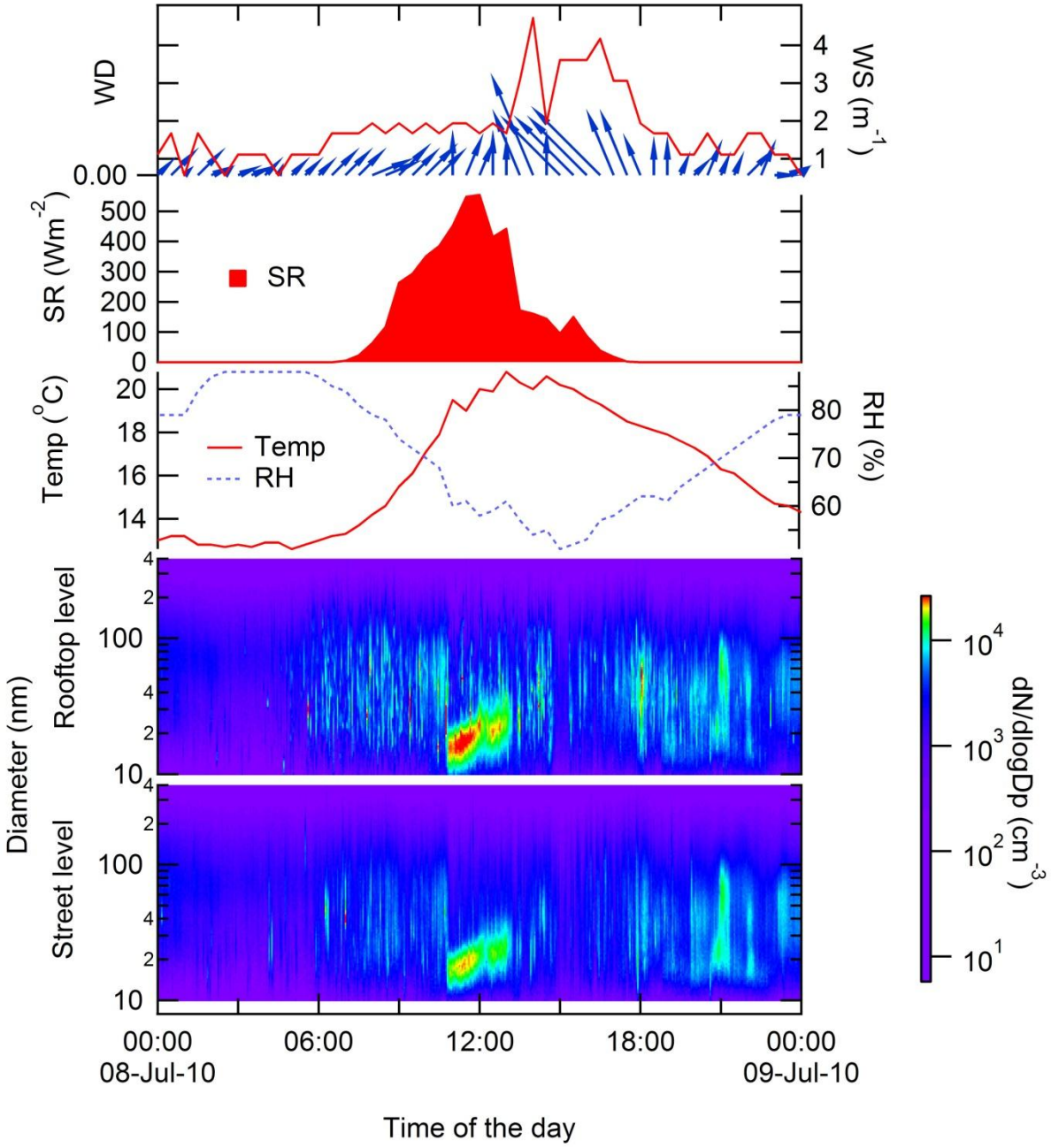

Fig. S9. PNSD spectra at Building C on a nucleation event day.