EXPERIMENTAL INVESTIGATION OF ULTRA FINE PARTICLE SIZE DISTRIBUTION NEAR A BUSY ROAD.

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ABSTRACT

In this paper, evolution of the size distribution of fine and ultra-fine particles emitted from vehicles on a busy road is investigated experimentally. In particular, several different modes (distinct maxima) of the particle size distribution are investigated as a function of the distance from the road. An unusual and complex evolution of these modes is discovered. In particular, it is demonstrated that some of the modes tend to shift towards smaller particle size, which has never been observed previously. Complex correlation between the modes is determined experimentally. In addition, for the first time, the total number concentration is clearly shown to go through a significant maximum at some distance from the road. This could have significant influence on the assessment of exposure to fine and ultrafine particles in the vicinity of busy roads.

Key words: Ultra fine particles; Size distribution; Busy road; Particle formation; Aerosol evolution.

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1. Introduction.

Fine and ultra-fine particles usually constitute only a small part of the overall particulate mass in an aerosol generated by pollution sources such as busy roads, and due to this reason until recently they were neglected by the Environmental Pollution Authorities. However, during the last several years, environmental and medical studies have revealed significant health risks related to fine and ultra-fine particle pollution from motor vehicles (Anderson et al, 1992, Dockery et al, 1993, Schwartz et al, 1996, Speizer, 1993). Supposedly, this is due to the ability of such particles to penetrate deeper into the respiratory tract and stay there for longer. This is why the latest research of environmentalists is focused on the following two problems. First, the investigation of the decay of the total number concentration of particles with distance from a busy road (Hitchins et al, 2000, Gramotnev et al, 2003). Second, the analysis of the modification of these particles as they are transported in the atmosphere (Alam et al, 2003, Zhu et al, 2002a, 2002b, Shi et al, 1999).

Shi et al, (1999) measured the size distribution for particles in the range from 10 nm to 352 nm at three distances downwind from a busy road. As a result, an insignificant shift (of only ~ 1 nm) of the size distribution was observed. However, these measurements were conducted in an urban area with a number of other roads in the vicinity of the measurement site, which made it difficult to make conclusions about aerosol evolution with increasing distance from the specific source (road). In another paper from the same group (Alam et al, 2003), unusual strong bursts of concentration of ultra-fine particles in the range ≤ 7 nm were registered at a significant distance from the road. These maximums were associated with the traffic conditions and solar radiation. An explanation of the obtained results by means of particle nuclea-

tion was attempted. However, the measurements were not specifically focused on the investigation of particle size distribution and its evolution with increasing distance from a busy road.

The first such an attempt has been undertaken in the two papers by Zhu et al (2002a,b), where the size distribution of particles in the range from 6 nm to 220 nm was investigated at six distances from a busy road: 17 m, 20 m, 30 m, 90 m, 150 m, and 300 m. Concentrations of particles were normalized to wind speed and direction (Zhu et al, 2002a), and averaged over different conditions during many days of observations. This research revealed an essential change in the size distribution of fine and ultra-fine particles during their transport from the road. A conclusion has been drawn that both coagulation of particles and turbulent dispersion contribute to the rapid decrease in particle number concentration (Zhu et al, 2002a). The process of coagulation was regarded as the main reason for the variation of the size distribution with distance from a busy road. Two main modes (strong maximums) of the size distribution (around ~ 10 nm and ~ 20 nm) that were observed at 17 m distance from the road, were claimed to shift towards larger particle diameters as the aerosol is transported away from the road (Zhu et al, 2002a). As mentioned above, concentrations were averaged over the atmospheric conditions and normalised to wind speed and direction. Therefore, this approach is very important for the determination of the general average tendencies of the aerosol evolution from a busy road. However, it may mask effects that are sensitive to particular atmospheric conditions and wind direction and speed. In addition, relatively large steps in distance might have resulted in insufficient spatial and temporal resolution of the measurements. For example, the assumption that the 20 nm mode shifts into the 30 nm mode (with increasing distance from 17 m to 30 m), and then into the 50 nm mode (at 90 m) due to particle coagulation may be seen as not sufficiently justified by the presented experimental data. Indeed, the curve corresponding to the 20 m distance in Fig. 4 from (Zhu et al, 2002a) suggests the opposite – the 20 nm mode actually shifts to the left (away from the 30 nm mode).

Therefore, the aim of this paper is in detailed experimental investigation of the evolution of ultrafine aerosol size distribution near a busy road. Several observed modes of the size distribution will be analysed. In particular, we will show that some of the modes (e.g., the 30 nm mode and 20 nm mode) tend to shift towards smaller particle diameters (contrary to what was previously suggested by Zhu et al (2002)). For the first time, clear evidence of an increase of the total particle number concentration at some distance from the road will be presented. Strong and unusual correlation between different modes of the size distribution will also be demonstrated.

2. Experimental procedure.

The experimental measurements were taken near the Gateway Motorway in the Brisbane area, Australia. The analyzed four-lane road (of the total width 27 m) and the surrounding area are presented in Fig. 1. The height of the Motorway above the surrounding ground level is ≈ 2 m. There are no buildings around the measurement area that is practically a flat grass field with isolated scattered bushes and trees. On the other (upwind) side of the Motorway, there is a small residential area with a parkland (Fig. 1).

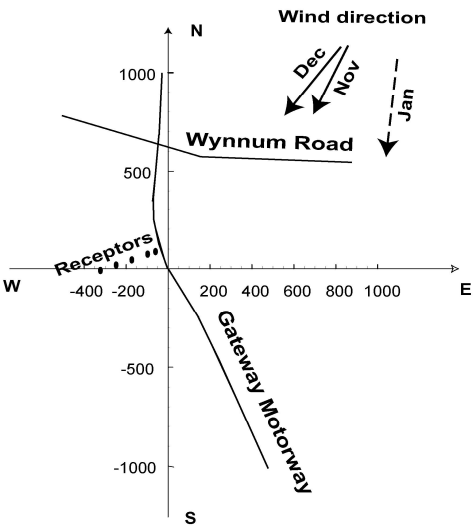

Fig. 1. Gateway Motorway with a few examples of receptor points. The wind direction is indicated by the solid arrows (for 20 November 2002 and 23 December 2002) and dashed arrow (for 8 January

2003). The scale of the map and the direction to the North are as indicated. The insert presents a section of the map of the area of measurements.

The total number concentration and size distribution of fine and ultra-fine particles in the range from 4 nm to 710 nm was measured at the height h = 2 m above the ground level (i.e. approximately at the level of the Motorway). Number concentration and size distribution of particles were measured by means of two scanning mobility particle sizers (SMPS) in two size ranges: from 4 nm to 163 nm in 100 equal intervals of $\Delta\log(Dp)$ (where Dp is particle diameter in nanometres), and from 14 nm to 710 nm in 110 equal intervals of $\Delta\log(Dp)$. Concentrations of particles within the larger range were measured by means of SMPS 3934 (the differential mobility analyzer DMA 3071A and the condensation particle counter CPC 3010). Concentrations of particles in the smaller range were measured using SMPS 3936 (model 3080 classifier with nano DMA 3085 and CPC 3025). The time for one full scan was 2.5 minutes. An automatic weather station was used to measure temperature, humidity, wind speed, wind direction and solar radiation at the same height h = 2 m and ~ 45 m from the middle of the road every 20 sec during the whole period of the concentration measurements.

The measurements were conducted on four different days: 20 November 2002, 23 December 2002, 8 January 2003 (weekdays), and 24 November 2002 (weekend). On 20 November 2002, both SMPSes were used simultaneously, while other measurements were taken only by means of an SMPS with the smaller size range from 4 nm to 163 nm. The concentration and size distribution measurements were taken at various distances from the road with increments from 6 m to 25 m within the range between 25 m and 307 m from the centre of the road. At each distance from the road, five consecutive measurement of the size distribution were conducted, from which the average size distribution was determined (see the figures in the next section).

The concentration and size distribution measurements were conducted simultaneously with the traffic flow. All the measurements were taken at approximately the same time intervals (from 11 am to 3 pm), when the traffic conditions on the Motorway were stable and variations of meteorological parameters were small (see below)(what to see below?).

Note that the main idea of the designed experiment was to determine the main features of the aerosol evolution at given (and sufficiently constant) meteorological and traffic parameters, such as humidity, temperature, wind speed, etc. This was the reason for taking the measurements at different distances from the road within a relatively short period of time (within several hours) when these parameters are approximately constant. The validity of such a method is clearly confirmed by the high level of confidence of all predicted features of the particle mode evolution (see Section 4).

3. Experimental results and discussion.

The average values of the meteorological and traffic parameters and their standard deviations are presented in Table 1.

Meteorological parameters	November 20	November 24	December 23	January 8
Wind direction (degrees to the North)	24 (±40)	25 (±45)	37 (±33)	7 (±27)
Wind speed (ms^{-1})	2.05 (±0.9)	2.2 (±0.8)	2.3 (±0.8)	2.6 (±1.0)
Normal wind component (ms^{-1})	1.15 (±0.5)	1.33 (±0.5)	2.05 (±0.7)	0.96 (±0.4)
Temperature (°C)	26.5 (±0.9)	27.1 (±0.4)	29.4 (±0.5)	28 (±1)
Humidity (%)	35 (±3)	36.7 (±0.9)	36 (±3)	42 (±4)
Solar radiation (W/m^2)	800 (±300)	1000 (±200)	900 (±200)	800 (±400)
Traffic (vehicles per hour)	3900	3700	5000	4300
Number of trucks (%)	20	3	23 (morning) 10 (afternoon)	16

Table 1. Average meteorological and traffic conditions

On 8 January, the wind was almost parallel to the road (the dashed arrow in Fig. 1), while on other days it had a noticeable component normal to the road (see Table 1 and the solid arrows in Fig. 1). In addition, the wind speed during all the measurements was relatively small (Table 1).

The typical average (out of five measurements) size distributions for various distances within the range from 25 m to 307 m from the centre of the road are plotted in Figs. 2 – 4 for 20 November 2002. The Friedman super smoother method (Venables & Ripley, 2000) was used for plotting the corresponding curves.

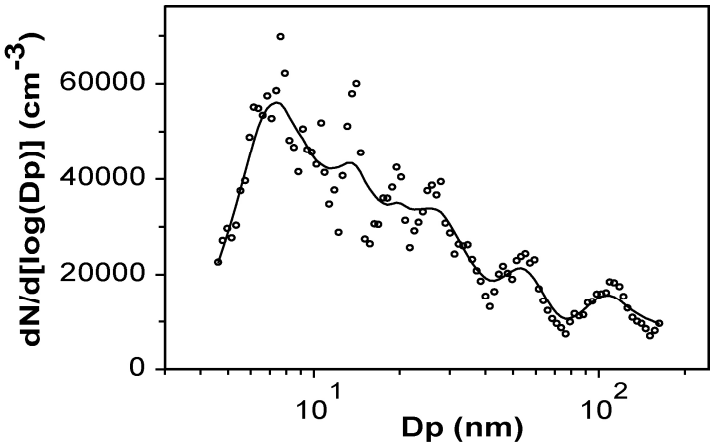

Fig. 2. The typical size distribution in the immediate proximity to the road (12 m from the kerb). The measurements were taken on 20 November 2002. The curve was plotted by means of the Friedman super smoother (Venables and Ripley, 2000).

Fig. 2 presents the experimental results for the average (over five measurements) particle concentrations in different channels at the distance 12 m from the kerb (in the upper paragraph the distance is referenced with respect to the centre of the road while here it is from the kerb. I think you should put all distances as from the center of the road), as a function of particle diameter. The super smoother curve reveals all the major, statistically significant features of the size distribution, i.e. maximums and minimums. Note that the height of the maximums on the super smoother curve (especially in the range from ~ 10 nm to ~ 30 nm) is no-

ticeably smaller than what is suggested by the experimental points (these maximums are statistically smoothed out).

It can be seen that six different maximums (particle modes) can be seen in the presented size distribution at about 7 nm, 12 nm, 20 nm, 30 nm, 50 nm, and 100 nm (Fig.2). All these modes, except probably for the 7 nm mode, were previously observed by different researchers. For example, the 12 nm, 20 nm, 30 nm, and 50 nm modes were reported previously in (Zhu et al, 2002a). It is important to note that the 7 nm mode is the strongest at the considered distance from the road. Further evolution of all these modes as the aerosol is transported by the wind away from the road is presented in Figs. 3 and 4.

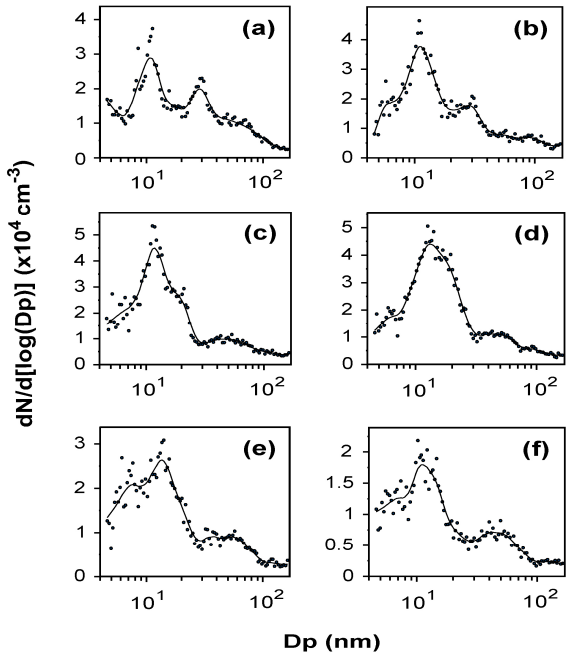

Fig. 3. The particle size distributions with the experimental values of the average concentrations in each of the 100 channels on 20 November 2002 (midweek) at the following distances from the centre of the road: (a) 45 m, (b) 57 m, (c) 70 m, (d) 82 m, (e) 107 m, (f) 132 m. The meteorological and traffic parameters, as well as their standard deviations, are presented in Table 1.

For example, Fig. 3 presents the experimental results for the particle size distributions at six distances from the road. Several distinct particle modes can be observed in this figure. The small dispersion of the experimental points clearly demonstrates the validity of the

smoothing technique, with all of the particle modes being statistically significant (for more detail see Section 4).

For the convenience of comparison of the presented dependencies and clear demonstration of the major features of mode evolution, the obtained smoothed dependencies of particle size distributions are re-plotted in the two figures 4a and 4b without the experimental points. As a result, the four distinct modes at the particle diameters ~7 nm, ~12 nm, ~30 nm, and ~50 nm are displayed by the curves in Figs. 4a,b. As can be seen from Figs. 4a,b, the 12 nm mode is fairly stable in position, and shifts only insignificantly with changing distance from the road (generally, this is in agreement with the results of Zhu et al (2002a)). However, what is more important, is that not too far from the road this maximum clearly increases in height with distance (curves 1 – 3 in Fig. 4a), despite the fact that dispersion must result in the opposite tendency. As far as we know, this result has not been described in the literature, and it is contrary to what was indicated previously in (Zhu et al, 2002a).

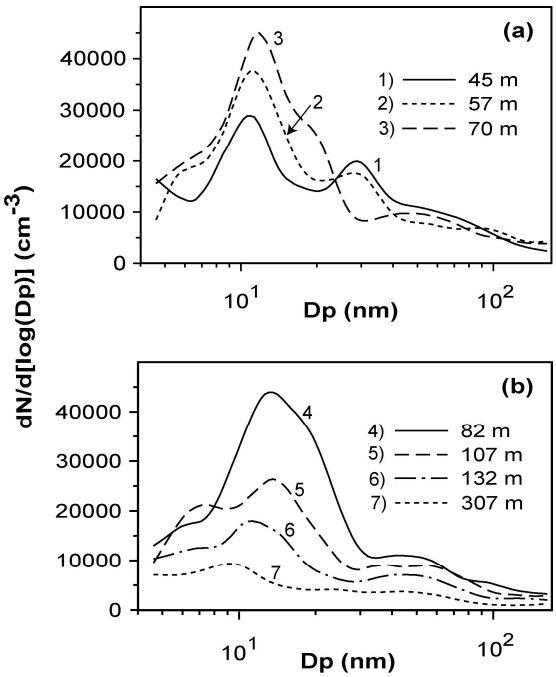

Fig. 4. The same size distributions as in Fig. 3 (re-drawn for comparison) without the experimental points.

Even more unusual behaviour is displayed by the 30 nm mode – see curve 1 in Fig. 4a. As has been mentioned in the Introduction, this maximum has also been observed by Zhu et al (2002a). However, their assumption that this maximum shifts noticeably to the right towards larger particle size (due to coagulation) with increasing distance from the road has not been confirmed by our results. For example, as demonstrated by curves 1 – 3 in Fig. 4a, the 30 nm mode slightly decreases in height with increasing distance from 45 m to 57 m (curves 1, 2), and then, at 70 m, noticeably shifts to the left rather than to the right (curve 3). This is indicated by a noticeable ”shoulder” (at ~ 20 nm) on the right of the 12 nm maximum (curve 3). A significant dip is left in place of the 30 nm mode. Further increasing distance to 82 m (curve 4, Fig. 4b) causes further increase of the 20 nm mode and its merger with the 12 nm mode, which results in a single broad maximum of approximately the same height as the 12 nm mode on curve 3. After this, increasing distance results in a monotonic decrease of both modes (curves 5 – 7, Fig. 4b).

At the same time, on the left of the 12 nm mode, there appears another maximum at ~ 7 nm (curve 5 in Fig. 4b). This is the same maximum as the main maximum in Fig. 2. The comparison of Fig. 2 with curve 1 in Fig. 4a (the next measured size distribution from the road) suggests that within the considered interval of distances, processes of particle formation (coagulation) have occurred, resulting in a significant decrease of the 7 nm mode in Fig. 2. Indeed, according to the Gaussian plume dispersion (Gramotnev et al, 2003), the decrease of concentration within the distance interval from 25 m to 45 m from the road (corresponding to the curve in Fig. 2 and curve 1 in Fig. 4a) must only be ≈ 1.8 times. However, the decrease of concentration of the 7 nm particles within the same distance interval appeared to be ≈ 3 times (even despite the possibility of an additional mechanism of particle formation due to nucleation). This suggests that there was a significant additional outflow (or sink) of particles from this mode, possibly due to particle coagulation.

The last obvious mode in Figs. 3 – 4 is represented by the maximum at ~ 50 nm that can be seen at distances ≥ 60 m (curves 3 – 6 in Fig. 4a,b). This is the mode that was interpreted by Zhu et al, (2002a) as the original 30 nm mode (curve 3) shifted substantially to the right due to particle coagulation. However, as has been demonstrated, this is not the case, since the 30 nm mode shifts in the opposite direction (curves 1 – 4 in Figs. 4a,b).

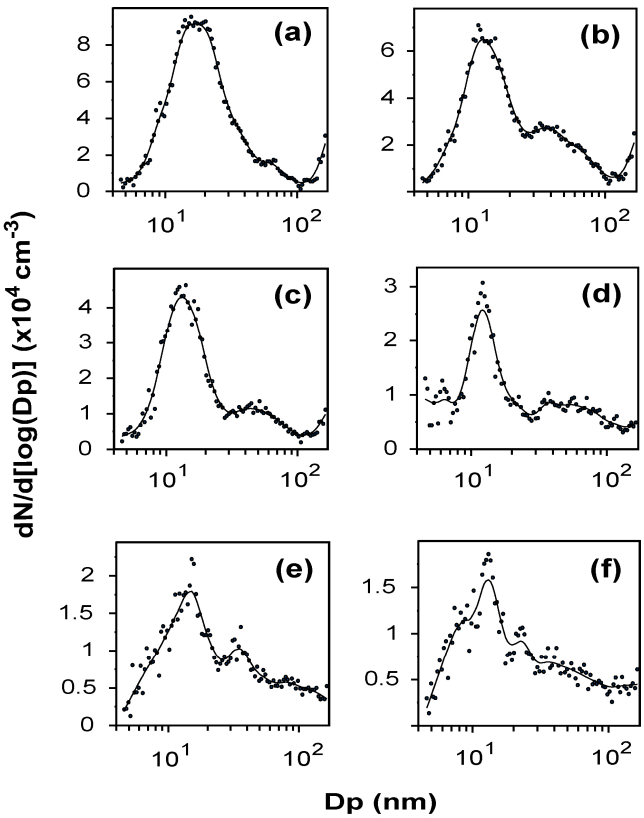

Fig. 5. The particle size distributions with the experimental values of the average concentrations on 23 December 2002 (midweek) at the following distances from the centre of the road: (a) 45 m, (b) 57 m, (c) 64 m, (d) 77 m, (e) 89 m, (f) 95 m. The meteorological and traffic parameters are presented in Table 1. Graphs (d) to (f) were measured in the afternoon when the number of heavy truck was significantly lower (see Table 1).

In order to confirm these fairly unexpected results, two other sets of measurements have been undertaken. Very similar behaviour of the size distribution modes has been observed on 23 December 2002 (Fig. 5 – 6). The normal component of the wind velocity in this case was noticeably larger than that on 20 November 2002 (Table 1). As a result, the 30 nm mode has appeared at significantly larger distances (~ 89 m) from the road – Fig. 5d. Other-

wise, the behaviour of this mode on 23 December is similar to that on 20 November (compare curves 1 – 3 in Fig. 4a with curves 5 – 6 in Fig. 6b). Very similarly, the mode at ~ 7 nm can also be noticed in Fig. 6b (compare curve 7 from Fig. 6b with curve 2 from Fig. 4a).

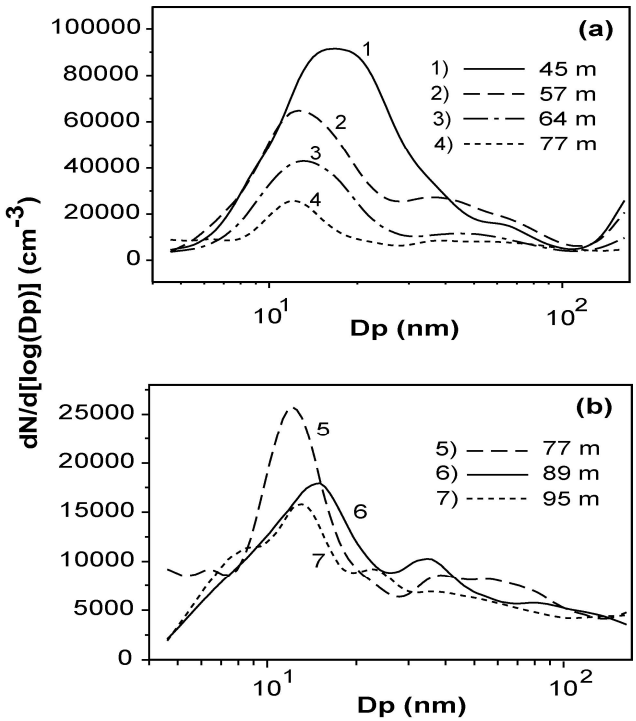

Fig. 6. The same size distributions as in Fig. 5 (re-drawn for comparison) without the experimental points.

The overall noticeably larger concentrations in Fig. 6a than those in Fig. 4a are due to the larger traffic flow, especially at the beginning of the experiment when curves 1 and 2 in Fig. 6a were obtained.

The observed increase of the 12 nm and 20 nm modes within the range between 45 m and 82 m (Fig. 4a) is expected to affect the behaviour of the total number concentration as a function of distance from the road. As a result, a substantial increase of the total number concentration at these distances from the road was observed on 20 November 2002 (squares in Fig. 7a). The maximum of the total number concentration is achieved at ≈ 82 m from the middle of the road (Fig. 7a). The error bars correspond to the standard deviation of the mean values of the total number concentrations, and clearly confirm that the considered maximum is statistically significant.

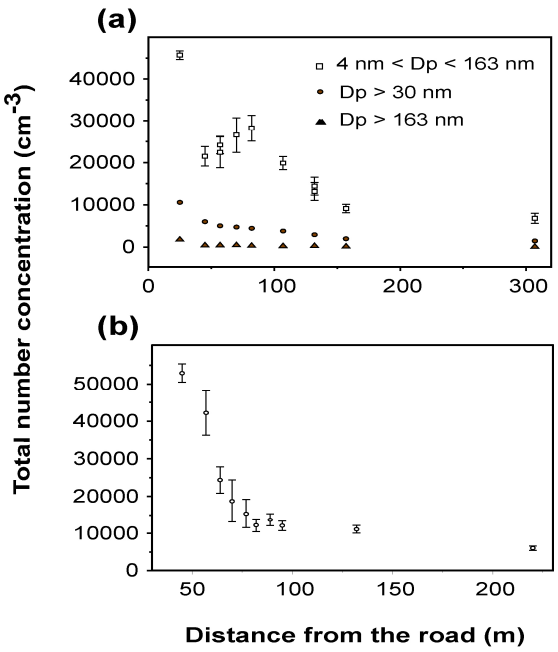

Fig. 7. The dependencies of the total number concentration of particles on distance from the road for the measurements on (a) 20 November 2002, and (b) 23 December 2002. The ranges of particles are as indicated. The error bars show the standard deviation of the average (over five measurements) total number concentrations. The double points at the distances 57 m and 132 m (in Fig. 7a) indicate the control sets measurements that were conducted on the end of the experiment to confirme the reliability and repeatability of the obtained results.

The dispersion of larger particles (> 163 nm) takes place in the usual fashion, i.e., results in a monotonic decay with increasing distance (triangles in Fig. 7a). Note that the overall concentration of these larger particles is negligible compared to the concentration within the range < 163 nm (Fig. 7a). Therefore, these larger particles do not have any effect on the observed maximum of the total number concentration. Moreover, as can be seen (dots in Fig. 7a), the total number concentration within the range > 30 nm also decays monotonically with distance, which strongly suggests that the observed maximum of the total number concentration is caused by some physical/chemical mechanisms resulting in increasing number of particles within the range ≤ 30 nm (for a detailed description of such mechanisms see (Gramotnev, submitted)).

The two additional points at the distances 50 m and 125 m (Fig. 7a) represent two control measurements of the total number concentrations at the end of the experiment. This was done to successfully confirm the reproducibility of the obtained results.

Note that the maximum of the total number concentration has also been observed on 23 December 2002 (Fig. 7b). This maximum appeared at the distance of ~ 89 m from the road, corresponding to the appearance of the 30 nm mode (see Fig. 4b). However, this maximum is much smaller than that in Fig. 7a, due to smaller particle concentrations. Nevertheless, the statistical t-test for comparison of mean values (Larsen and Marx, 1986) has demonstrated that the probability of existence of this maximum is more than 70% (compared to 94% for the similar maximum in Fig. 7a).

The obtained results and dependencies are strongly affected by the type of vehicles on the road. For example, measurements on weekend (24 November 2002) have resulted in noticeably different dependencies – Fig. 8. In particular, the 12 nm maximum is relatively small (Fig. 8), which is related to the significantly smaller number of heavy duty diesel trucks in the traffic flow. This is in agreement with the earlier conclusion that this mode is mainly due to diesel vehicles (Zhu et al, 2002a). Otherwise, the dependencies in Figs. 4a,b, 6b and 8 are fairly similar.

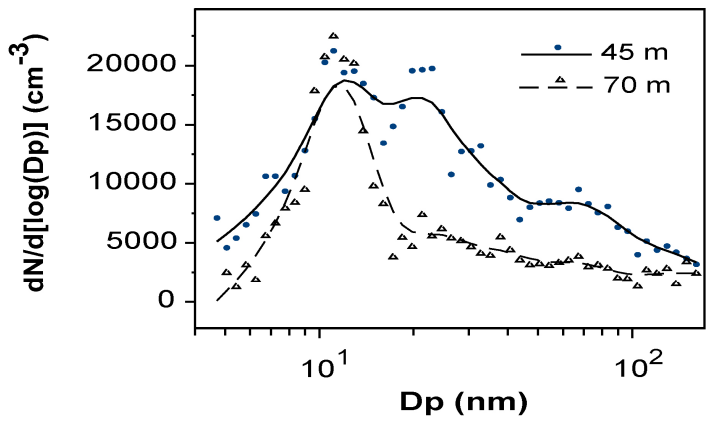

Fig. 8. Particle size distributions on 24 November 2002 (Sunday), with significantly lower contribution of trucks (see Table 1).

The measurements on 8 January 2003 have resulted in a substantial scatter of concentrations within channels, not allowing repeatable size distributions. This is due to the wind being almost parallel to the road (Table 1 and Fig. 1). In this case, only slow transport of the aerosol from the road occurs, causing larger particle concentrations. On the other hand, large tangential component of the wind results in increased turbulence and, therefore, fluctuations of concentration. Thus, the evolution effects are masked by strong turbulent fluctuations. In this circumstances, significantly more than five measurements of size distribution are required to obtain a repeatable average size distribution.

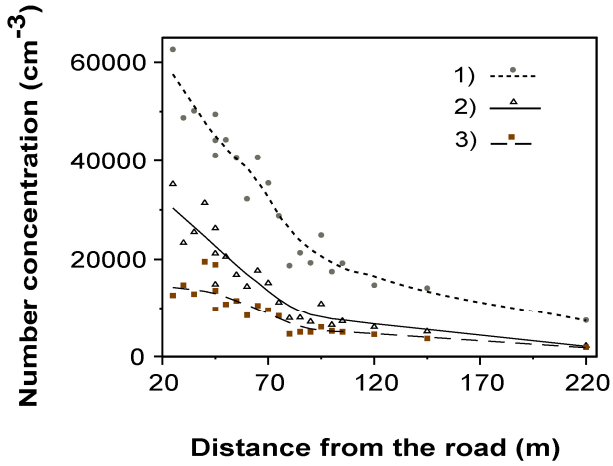

Fig. 9. Total number concentrations on 8 January 2003 (almost parallel wind) within the particle ranges: 1) from 4.6 nm to 163 nm; 2) from 7 nm to 20 nm; 3) from 21 nm to 42 nm (includes the 30 nm mode). The curves are plotted by the Loess regression (Venables and Ripley, 2000).

The dependence of the total number concentration on distance from the road for 8 January 2003 is shown in Fig. 9 by the full circles for the range of particles from 4 nm to 163 nm. In this case, methods of simple curve fitting based on the exponential function $c = k_1 e^{-\alpha d}$ (Hitchins et al, 2001, and Zhu et al, 2002a) or the power law $c = k_2 d^{-m}$ (Gramotnev et al, 2003) with $k_1 = 7.0\times10^4$, $\alpha = 0.015$, and $k_2 = 2\times10^5$, $m = 0.5$ can be used. The t-values (Larsen & Marx, 1986) in these cases are as high as 17, 10, 5, and 8 for k_1, α, k_2, and m, respectively. This indicates that the level of confidence for fitting both exponential and power curves exceeds 99%.

On the other hand, in order to reveal small features of the dependencies of the total number concentration on 8 January 2003, the Loess regression (Venables & Ripley, 2000) was used. This method is more efficient (than the Friedman super smoother) in drawing regression curves with small features represented by a few experimental points (Venables & Ripley, 2000). The obtained regression curves are shown in Fig. 9 for the total number concentrations within the three size ranges as functions of distance from the road. In particular, it can be seen that curve 3 corresponding to the 30 nm mode displays a tendency to levelling at the distances between 25 m to 70 m. This indicates an inflow of particles into this mode. This is probably the reason for the small bump on curve 1 at ~ 50 m from the road. As a result, the effects of the mode evolution observed in Figs. 3 – 8 has only a very limited effect in the case of almost parallel wind.

4. Level of confidence and errors of the obtained results

There are several sources of possible errors in the presented experimental results. In this section, we will consider their contribution to the level of confidence of the obtained features of mode evolution and will demonstrate the validity of the obtained results.

The first source of errors can be associated with changing atmospheric parameters. However, as shown in Table 1, standard deviations of such vital parameters as humidity and temperature are small during each of the measurement periods. These small variations of humidity and/or temperature are unlikely to affect the presented curves for size distributions. This, however, does not mean that temperature and humidity do not affect the described mode evolution. On the contrary, they are expected to be crucial for understanding the behaviour of the modes – see (Gramotnev (submitted)).

The standard deviations of speed, direction, and normal component of the wind are large (as expected) due to strong turbulence (Table 1). However, the associated random errors of particle concentrations in this case are effectively reduced by taking the average of five

scans at every distance from the road (see Section 2). This statement is clearly confirmed by the limited dispersion of experimental (average) points around the size distribution curves (Figs. 3 and 5).

Solar radiation changed fairly noticeably (Table 1). However, no direct and obvious effect of solar radiation on the observed mode evolution has been detected. How do you know this. We did not present any evidence that the solar radiation did not have any influence. Once again, this does not mean that there is no such an effect (which should be the matter for further investigation), but rather that the appearance and transformation of the modes in our experiment did not correlate with the solar radiation.

Standard deviations of the overall traffic (for the traffic flow measured in 5 minute intervals) on all days of measurements were less than 10%, and the standard deviations for trucks ~ 20%. This is a clear indication of a highly stable traffic conditions on the considered road. Insignificant contribution of traffic variations to the errors of the measured particle size distributions and the total number concentrations is also demonstrated by the control measurements conducted in the end of the measurement period on 20 November 2003 (see the additional two points at the distances 57 m and 132 m in Fig. 7a). Not only was the observed total concentration in the repeated measurements the same but also the two measured size distributions at the distance 57 m for the original (curve 2 in Fig. 4a) and control sets of five scans were not significantly (can you say statistically not significantly different) different. This is also another confirmation of the high stability of the obtained results with respect to the fluctuations of the meteorological parameters, including wind speed and direction.

At the same time, it is important that on 23 December 2003, traffic conditions changed substantially for the morning and afternoon measurements (due to the next day holiday). The number of trucks in the afternoon has dropped very noticeably (see Table 1). This was one of the major reasons why the 30 nm mode and 20 nm modes in Fig. 6b were substantially smaller than those in Fig. 4a. We also believe that this is the reason for the maximum of the

total number concentration in Fig. 7b to be significantly smaller than in Fig. 7a. Nevertheless, despite such a drastic variation in the traffic flow, the major features of the mode evolution have remained unchanged (compare Fig. 4a with Fig. 6b). This clearly demonstrates that the discovered unusual features of the aerosol evolution are highly resilient to variations of traffic flow, and that they are not an artefact of changing atmospheric and traffic conditions.

In addition, levels of confidence for all of the maximums of the size distributions (modes) in Figs. 4, and 6 were determined by means of known statistical methods (give the name of the statistical method (Bowman and Azzalini, 1997). Using the S-Plus package, the experimental point near each of the modes are approximated by a polynomial of the fifth order (to include the neighbouring modes). The level of confidence of the considered mode is then determined using the two neighbouring minima and the error of the approximation (Bowman and Azzalini, 1997). The resultant values of the level of confidence for the modes in Figs. 2 – 6 are presented in Tables 2 and 3, clearly demonstrating the validity of the obtained results and conclusions.

	25 m	45 m	57 m	70 m	82 m	107 m	132 m	307 m
7 nm	>99	-	-	-	85	91	-	>99
12 nm	83	>99	>99	>99	>99	>99	>99	-
20 nm	83	-	-	-	-	-	-	-
27 nm	>99	>99	>99	-	-	-	-	-
50 nm	>99	-	-	99	81	91	>99	79

Table 2. Levels of confidence in % for different modes on the curves in Figs. 2 – 4 (20 November 2003). The modes are shown in the first column, while the distance from the road (corresponding to different curves in Figs. 2 – 4) are shown in the first row.

	45 m	57 m	64 m	77 m	89 m	95 m
12 nm	>99	>99	>99	>99	>99	>99
20 nm	-	-	-	-	-	89
30 nm	-	-	-	-	>99	-
40 – 50 nm	-	>99	99	>99	-	-

Table 3. Levels of confidence in % for different modes on the curves in Figs. 5 – 6 (23 December 2003).

Finally, it can be shown that the effect of Wynnum road (Fig. 1) is negligible. Indeed, the Gaussian plume approximation gives that particle concentrations should decay as a power function of distance from the road (Gramotnev et al, 2003). Taking into account the typical traffic flow on Wynnum road (2127 vehicle/hour with 1.8% of heavy duty trucks), the distance to the place of measurements (600 – 800 m), and the average emission factors for light cars and heavy-duty trucks (Gramotnev et al, submitted), we obtain that the typical concentrations due to Wynnum road are at least ~ 10 times less than those from the Gateway Motorway at all sampling points.

5. Conclusions

In this paper, for the first time, a detailed experimental investigation of the evolution of particle size distribution modes in the fine and ultra-fine ranges has been carried out near a busy road. A number of unexpected and unusual effects have been observed. These are related to mutual transformation of different modes, resulting in appearance, growth and disappearance of these modes at particular distances from the road. For example, the first clear and consistent experimental evidence of the existance of a strong 7 nm mode of particles has been obtained. A strong mode at ~ 30 nm particle diameter appears at a particular distance from the road (why did you remove this sentence?). This mode has been shown to consistently shift to the left, i.e. towards smaller particle diameters. As a result, the 30 nm mode is transformed into the 20 nm mode, and then merged into the 12 nm mode. At the same time, it has been shown that during the whole process of mode conversion and transformation, the particle concentration in the 7 nm mode changes only insignificantly (increasing at some stages) with increasing distance from the road. Therefore, since dispersion must obviously result in a signifi-

cant reduction of particle concentrations, there is a substantial influx of particles into the 7 nm mode during the aerosol evolution and its transport away from the road.

For the first time, clear experimental evidence of a maximum of the total number concentration at a particular distance from the road has been obtained. This maximum has been shown to occur due to a strong increase of particle concentration within the range < 30 nm. The observed maximum has been demonstrated to decrease and move away from the road with increasing normal (to the road) component of the wind. The effect of normal wind component on the particle size distribution has also been investigated. For example, increasing normal component of the wind results in approximately proportional increase of the distance at which the 30 nm mode appears. If the average wind is approximately parallel to the road, then the observed modes are shown to significantly fluctuate due to the turbulence and stochastic nature of fluctuations of wind direction. The existence of a maximum of the total number concentration at a particular distance from the road can have a significant influence on the assessment of the exposure to fine and ultrafine particles in the vicinity of the road.

Statistical analyses of the errors associated with the experiments have been conducted, and the levels of confidence for the observed modes of particle size distribution have been determined, clearly confirming the obtained results and conclusions.

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