1 **Ground-fixed and on-board measurements of nanoparticles in the wake of a moving vehicle**

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6 **Abstract**

7 An integrated experimental methodology has been applied to measure number and size distributions
8 of particles in the 5–560 nm size range in the wake of a diesel car running at different speeds.
9 Measurements were made at both ground-fixed (0.10 and 0.25 m above the ground level) and on-
10 board (in 12 different sampling locations behind the moving car) measurement configurations using
11 a fast response differential mobility spectrometer (Cambustion DMS50) with a sampling frequency
12 up to 10 Hz. Results from both the experimental campaigns were analysed to understand the
13 dynamics, dispersion and transport of nanoparticle emissions in the wake of a moving vehicle.
14 Temporal changes in results were divided into three main stages (pre-evolution, evolution and post-
15 evolution) after the release of exhaust emissions from the tailpipe. Evolution stage is of most
16 interest where all the changes to particle number and size distribution occurred. Up to four
17 evolution sub-stages were observed, each showing distinct evolution patterns of particle size
18 distributions, depending on the particular experimental run. In agreement with previous studies,
19 dilution was found to be the dominant process throughout all the evolution stages. The first
20 evolution sub-stage was common to all the measurements, and consisted of an initial particle
21 number concentrations and distributions change due to rapid (less than 1 s) nucleation followed by a
22 rapid increase of accumulation mode particle number concentrations. After this first sub-stage the
23 presence of vehicle wake with recirculating particles and the possible influence of other
24 transformation processes lead to complex interactions. Results from the two experimental data sets
25 clearly confirm the presence of two separate groups of particles: (i) new particles, which are freshly
26 emitted and come directly from the tailpipe and (ii) relatively aged particles, which are entrained
27 within the recirculation vortices of the vehicle wake and reside there for a longer time. The two
28 groups have different characteristics and interact with each other. This interaction has often been
29 overlooked in past studies about local scale dispersion of nanoparticle from moving vehicles.

30 **Keywords:** Fast response measurements; Nanoparticles dispersion; Number concentrations; Size
31 distribution; Ultrafine particles; Vehicle wake

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1. Introduction

Nanoparticles in the atmosphere are responsible for a number of adverse effects on human health and the environment (Davidson et al., 2005; Strawa et al., 2010). Road vehicles are the dominant source of particles by number (Kumar et al., 2011a). Particles below 300 nm (referring here as nanoparticles to describe particle number concentrations, PNCs, throughout this article) can contribute up to 99% of total PNC in the urban environment (Kumar et al., 2008b; Kumar et al., 2009a). Measuring and characterising emissions from individual vehicles under operational driving and dilution conditions is an important area of research. However, studying nanoparticle dispersion at fine spatial and temporal scales (e.g. in a vehicle wake) becomes a complex task, mainly because of the limited time response of most available instrumentation (Kumar et al., 2010). Our recent review summarised studies on field measurements and dispersion modelling of nanoparticles in vehicle wakes (Carpentieri et al., 2011). The review highlighted very short time scales associated with nanoparticle transformations in the earliest stages of emission. These transformations strongly interact with the flow and turbulence field immediately behind the emitting vehicle; hence, there is a need for characterising in detail the mixing processes in the wake, especially in the near wake.

A recent review by Kumar et al. (2010) reported that several field measurement studies of number and size distributions of nanoparticles in urban areas have been made using instruments such as scanning mobility particle sizers (SMPS), condensation particle counters (CPC), electrical low pressure impactors, alone or in combination. Most of these instruments have lower sampling frequencies relative to that required to characterise nanoparticle dispersion phenomena in vehicle wakes, hence an instrument with a fast response is required (Kumar et al., 2009c).

Field experiments purposely designed for measuring nanoparticle evolution in the wakes of vehicles are rare. Ambient measurements, while providing an integral view of the population exposure levels, cover both primary emitted and secondary formed particles, thus they cannot be reliably used to derive detailed source–receptor relationships (Wehner et al., 2009) or to investigate the first stages of the particle evolution within the wake (Carpentieri et al., 2011). Most of the previous

studies on nanoparticle emissions from single vehicles in real driving conditions (as opposed to direct laboratory measurements using chassis dynamometers) are based on on-road measurements with so-called chasing methods (Wehner et al., 2009). In these experiments, mobile laboratories equipped with measurement instruments follow the car to be investigated and measure the particle and gas emissions; for example, see Vogt et al. (2003), Giechaskiel et al. (2005) and Kittelson et al. (2006). The main limitation of these methods is the minimum distance between the two vehicles (i.e. chasing and experimental) which cannot be smaller than 10–15 m for safety reasons (Wehner et al., 2009), making the technique unsuitable to investigate nanoparticle transformations in the near wake, especially in the very first stages from emission.

New techniques have recently been developed to measure closer to the tailpipe. For instance, Morawska et al. (2007) used a plume capture trailer to sample the naturally diluted air within 1–3 m behind the tailpipe of a moving diesel car. However, the vehicle wake was heavily distorted due to the presence of the trailer and the samples were taken offline, making impossible a realistic analysis of the evolution process. Wehner et al. (2009) overcame this problem by using on-board measurements. They mounted samplers for measuring meteorological parameters (ambient temperature and relative humidity), PNCs in the 7–400 nm size range and gas (NO_x and CO_2) concentrations, upon a bicycle rack on the back of a moving car. Using this experimental setup, they measured number and size distributions of particles between 0.45 and 0.90 m away from the tailpipe and derived particle number emission factors under different driving conditions. This experimental setup reduced the disturbance on the wake flow generated by the mounted instrumentation to acceptable levels. However, the number of measuring devices deployed, while being very useful for the post-processing and interpretation phases still produced a non-negligible modification of the vehicle wake. The study used 90s averages from samples analysed by a SMPS; this could be further improved by using instruments with shorter response times and higher sampling frequencies.

A completely different approach was used by Hak et al. (2009). They used ground-fixed sampling

84 to characterise nanoparticle emissions from individual vehicles by means of a CPC at a fixed
85 location on the road surface. While capable of providing interesting information on individual
86 particle number emission factors, the long response time of the instrument ($T_{90} = 3$ s) and sampling
87 frequency (1 Hz) limited the detailed investigation of the exhaust plume. They found the duration of
88 the PNC peak to be on the order of a few seconds, as was also confirmed by the experiments carried
89 out by Kumar et al. (2009c). While integral information could be extracted with this setup, higher
90 frequencies and shorter response times are needed to capture the particle transformation processes
91 in the emitted plume. In order to overcome the latter problem, Kumar et al. (2009c) used for the
92 first time a fast response particle spectrometer to characterise the evolution of number and size
93 distributions of particles emitted by a diesel van with a sampling setup similar to the one described
94 above. Their preliminary measurements, obtained by using a Cambustion DMS500 with a 10 Hz
95 sampling frequency, highlighted the reliability of the measurement setup and allowed capture of the
96 nanoparticle evolution process in the vehicle wake with the required detail. Therefore, one of the
97 two parts of the methodology used in the present study involved the application of this technique
98 with a more systematic approach for studying the evolution of particle number and size distributions
99 in the wake of individual vehicles.

100 Considering the above, this study used an integrated measurement approach combining both
101 ground-fixed (Section 2.1) and on-board (Section 2.2) measurements together with a portable fast
102 response differential mobility spectrometer (Cambustion DMS50). The experimental setup allowed
103 capturing of detailed high spatial and temporal resolution data sets, while minimising the distortion
104 effects on the vehicle wake. Field measurements were conducted with the following objectives: (i)
105 application and validation of an systematic experimental methodology allowing fast response
106 measurements of PNCs while minimising vehicle wake distortion, (ii) understanding the dynamics,
107 dispersion and transport of particle emissions in the wake of moving vehicles, (iii) investigating
108 interaction between nanoparticle dilution and other transformation processes under various driving
109 conditions, and (iv) providing reference cases for future work, involving the interpretation of wind

110 tunnel experimental data and the validation of purposely developed mathematical models.

111 **2. Experimental setup and data collection**

112 **2.1. Ground-fixed sampling**

113 The experimental campaign was performed at the University of Surrey car park. Fig. 1
114 shows the schematic representation of the experimental setup. The first set of tests was performed in
115 a single lane of the car park about 70 m long and 6 m wide. The vehicle used for the experiments
116 was a University owned Vauxhall Astra Van, which has a 1686 cm³ diesel engine and was registered
117 in 2004 under the Euro 4 emission standards. Exhaust emissions were sampled with a 4 m long
118 sampling tube placed above the road surface at two fixed heights (0.10 and 0.25 m), connected to
119 the DMS50. The DMS50 has a sampling frequency up to 10 Hz, with a response time ($T_{10-90\%}$)
120 equal to 500 ms, and is capable of measuring number and size distributions in the 5–560 nm size
121 range. The instrument is controlled by a laptop connected via an Ethernet interface.

122 Intentionally, the experimental campaign was carried out during a weekend (Saturday, 16th October
123 2010) so that there was negligible vehicle movement near the site in the car park. To avoid any
124 influence of emissions from any external vehicle on our measurements, we made sure that no other
125 cars were moving near the site and the background PNCs were steady or re-established in case
126 there were emissions from other vehicles. Measurements were taken at both heights (i.e. 0.10 and
127 0.25m above the road surface) separately after driving the car at four different speeds representing
128 typical urban driving conditions (i.e. 20, 30, 40 and 50 km h⁻¹). Each set of experiment generated
129 from the cross-combination of experimental conditions (car speed and sampling height) was
130 repeated 6 to 10 times for reproducibility purposes (see Table 1). The car was driven above the
131 sampling point, which was distant, in the lateral direction, approximately 0.27±0.15 m from the
132 tailpipe exit (Fig. 1). The tailpipe had a diameter equal to 0.05 m, and its centre was approximately
133 0.30 m above the road surface.

134 Meteorological data (wind speed and direction, temperature and relative humidity) were monitored

135 locally by using two weather stations. The first one (Davis Weather Wizard 3) is located on top (~20
136 m above ground level) of a nearby University building which was about 250 m away from the
137 experimental site. This station measured the data at 2 s intervals and logged them every 10 minutes
138 after averaging. For monitoring the local meteorological conditions at the experimental site, the
139 second weather station (LaCrosse WS-2350) was placed on a pole at 4 m above the road level near
140 the sampling location (see Fig. 1). This station logged the monitored meteorological data every 3
141 minutes during the experiments. Weather conditions remained relatively constant during the
142 experimental campaign, with an average temperature of 15.9 °C, an average pressure of 102.1 kPa
143 and average relative humidity of 59%. The average wind speed measured by the campus weather
144 station on the building roof was 4.9 m s⁻¹ with a prevailing wind direction approximately from
145 north-east, while the on-site station measured an average of 1.4 m s⁻¹ at a much lower height.

146 **2.2. On-board measurements**

147 The second experimental campaign was performed again in the University of Surrey car
148 park. The same car, as described in Section 2.1, was driven around the perimeter lane of the car park
149 during two separate campaigns (Saturday, 13th November 2010, and Saturday, 29th January 2011).
150 Similar to ground-fixed measurements, three car speeds (20, 30, and 40 km h⁻¹) were used during
151 these experiments. The area used for these experiments (see Site 2 in Fig. 1) has an approximate
152 extension of 300 m in the east-west direction. This arrangement was sufficient for driving the car at
153 an approximately constant speed for about 20 to 60 s during each run in both directions. All the runs
154 were repeated at least twice.

155 In this second experimental campaign, the DMS50 was placed inside the test vehicle and the sample
156 tube (length 1.75 m) was attached to an L-shaped pole extending from the back of the car (see Fig.
157 1). The set up covered both the traditionally seen recirculating flow regions on each corner of a
158 moving vehicle and centre region (Hucho, 1987), and allowed mapping of number and size
159 distributions at 12 fixed points within the vehicle wake. These included 9 (3×3) points at $x=0.45$ m
160 (in particular at $y=-0.45$, 0 and 0.45 m, and $z=0.10$, 0.50 and 0.90 m), and 3 further points at $x=0.80$

161 m (at $y=-0.45$, 0 and 0.45 m, and $z=0.50$ m). Note that, as illustrated in Fig. 1, $x=0$ is placed at the
162 tailpipe exit, $y=0$ is located at the car centre line and $z=0$ is at ground level, so that $y=-0.45$ m is in
163 line with the tailpipe.

164 Meteorological conditions were monitored as described in Section 2.1. The first of the two on-
165 board experimental campaigns was carried out in a day with very calm wind conditions. The
166 average measured values during these tests were: temperature 11.4 °C; atmospheric pressure 99.5
167 kPa; relative humidity 64%; wind speed 0.4 m s⁻¹; prevailing wind direction from west. During the
168 second on-board campaign wind conditions were similar to those during the ground-fixed tests:
169 average wind speed at roof level equal to 4.8 m s⁻¹, with a prevailing wind direction from south;
170 average temperature, atmospheric pressure and relative humidity were, respectively, 3.9 °C, 102.6
171 kPa, and 62%.

172 3. Results and discussion

173 This chapter reports and discusses the results obtained from both (ground-fixed and on-
174 board) the experimental campaigns. After few preliminary checks for quality assurance of data
175 (Section 3.1), experimental results are presented in terms of PNCs and particle number distributions
176 (PNDs) in Sections 3.2 and 3.3 for both experimental setups. Discussion on the main findings and
177 significance of the experimental study is then reported in Section 3.4.

178 3.1. Sensitivity levels on the DMS50 and particle losses in sampling tubes

179 In order to ensure the quality of the measurements with the Cambustion DMS50, some
180 preliminary checks were deemed necessary. In particular, sensitivity levels compared with noise
181 was analysed, and an initial assessment of particle losses in sampling tubes was carried out.

182 Following the same methodology applied by Kumar et al. (2009c) for testing the noise level of the
183 DMS500 in their experiments, sensitivity of the DMS50 was checked by comparing its noise level
184 with the measured minimum (background) PNCs at the experimental site. The background PNCs at
185 site were measured by applying the highest sampling frequency (10 Hz) when the noise level of the

186 DMS50 is also the greatest. As seen in Fig. 2, comparison of the measured PNDs at a maximal
187 noise (10 Hz frequency) indicates that the background PNCs were well above the noise level of the
188 instrument for particle diameters greater than 7 nm. The signal from particle sizes up to 7 nm,
189 however, is comparable, or below, the noise level at 10 Hz. This fact, however, does not affect the
190 discussion on the following sections, since most of the analysis is based on longer time averages.

191 Particle losses caused by the experimental set up can become relevant especially for smaller particle
192 sizes and longer sampling tubes (Kumar et al., 2008c). In order to minimise this effect, the length of
193 the sampling tubes has been kept to the minimum required to reach the measurement location. As
194 described in Section 2, this length was 4 m for ground-fixed measurement, and 1.75 m for on-board
195 measurements. The internal diameter of the tubes is 5 mm. Sampling flow rate was kept to 0.00014
196 $\text{m}^3 \text{s}^{-1}$ with a line pressure of 80 kPa. The Reynolds number (Re) was around 1.7×10^3 , thus within
197 the laminar flow limit.

198 According to previous studies (Kumar et al., 2008c), we can expect that particle losses should
199 become relevant only for smaller sizes (< 20 nm), with the greatest effects in the 5–10 nm range.
200 Using the semi-empirical formulation given by Hinds (1999), for example, the penetration
201 efficiency during the tests with the 4 m tube can be estimated as around 70–75% for particle sizes $\leq$
202 6 nm, reaching already 80% at 7 nm, and above 90% for particles bigger than 10 nm. For the 1.75 m
203 long tube, penetration is above 80% for all size ranges detected by the DMS50, reaching 90% for
204 particle sizes around 7 nm.

205 In the following sections results are discussed mainly in relative terms, comparing data from
206 different runs, at different locations and time. In absolute terms, however, the main effects would
207 be: (i) underestimation of total PNCs and, in particular, of PNCs in the 5–30 nm size range, that are
208 often mentioned); (ii) overestimation of total geometric mean diameters (GMDs) and, in particular,
209 of GMDs in the 5–30 nm size range.

210 **3.2 Analysis of PNCs**

211 3.2.1 PNCs during ground-fixed measurements

212 As reported in Section 2.1, 8 different experimental cases were studied (height $H = 0.10$ and
213 0.25 m, with car speed $V=20, 30, 40$, and 50 km h^{-1}). Several runs were repeated within each
214 experimental set, i.e. 10 runs for car speeds up to 40 km h^{-1} , and 6 runs for 50 km h^{-1} ; the runs for
215 each experimental case are numbered sequentially from Run 01 to Run 10 (or 06 in the case of
216 $V=50 \text{ km h}^{-1}$), leading to a total of 72 datasets available for analysis from the ground-fixed
217 measurements. Seven runs were ignored in the post-processing stage due to quality issues caused
218 by external disturbances and measurement errors. A summary of the results from these
219 measurements is reported in Table 1.

220 Table 1 reports the average peak value of PNCs for each experimental case, along with its standard
221 deviation. While the average peak value of total PNC was within one order of magnitude (i.e. in the
222 $10^5\text{--}10^6 \text{ \# cm}^{-3}$ range) for all experimental cases, the actual value during each run varied within a
223 greater range, as evidenced also by the calculated standard deviations (Table 1). In particular, the
224 ratio between the maximum measured peak value ($H=0.25$ m, $V=50 \text{ km h}^{-1}$, Run 02) and the
225 minimum measured peak value ($H=0.10$ cm, $V=40 \text{ km h}^{-1}$, Run 04) was about 50. This variability,
226 even within the same nominal experimental conditions, was largely expected, given the
227 unpredictability of local turbulence conditions and imperfect alignment of the car with respect to the
228 sampling point across different runs (see further discussion on this point in Section 3.4).

229 Much more consistent is the time when the instrument measured the peak value for a particular run
230 (Table 1). Confirming results from our earlier work (Kumar et al., 2009c), the PNCs reached their
231 peak values within and just over 1 s for $H = 0.10$ and 0.25 m, respectively; $t=0$ is defined as the
232 time when the PNC start to significantly increase from the background concentrations which can
233 supposedly be considered as the first sign of detection of the plume. As expected, the time when the
234 peak is detected seems to be affected more by the sampling height than the vehicle speed, as
235 evidenced by the greater difference in peak times between 0.10 and 0.25 m than between cases at
236 different car speeds. The duration of the event, arbitrarily defined as the time during which the PNC

237 is above 150% of the background value, seems to increase on average together with the vehicle
238 speed for both $H = 0.10$ and 0.25 m. In particular, the decay time appears to increase with the car
239 speed, as reported in the following analysis.

240 Fig. 3 shows the temporal evolution of normalised average PNCs for each experimental case. These
241 normalised values are firstly obtained by dividing all the PNC values by the peak PNC value for
242 each individual run. Then, for each experimental case, values of different runs have been averaged
243 considering PNCs at the same time (defining $t=0$ as explained above). This average curve was then
244 re-normalised using the average peak value for each experimental case to obtain curves with peaks
245 equal to 1. The duration for the peak PNCs is generally larger for higher car speeds as seen in Table
246 1 and Fig.3. In particular, the decay time of the peak PNCs towards the site-background increases
247 with the increasing car speed. This increase in decay time with car speeds could be due to a
248 combination of two factors: (i) emission factors at higher car speeds are higher (Kumar et al.,
249 2011b), and (ii) the effects of the wake at the sampling location last longer.

250 As evidenced by the error bars shown in Fig. 3, the average normalised PNCs obtained for the first
251 three experimental cases ($H= 0.10$ m; $V = 20, 30$ and 40 km h⁻¹) have a remarkably low variability.
252 However, a much greater variability is observed, especially during the peak decay phase, both at
253 0.10 and 0.25 m sampling heights for $V = 50$ km h⁻¹. The variability for the other experimental
254 cases ($H = 0.25$ m, $V = 20, 30$ and 40 km h⁻¹) is between those two levels. In particular for $V = 20$
255 and 30 km h⁻¹, besides the variability in the decay phase, there is a smaller initial peak which is not
256 always present in all the runs, increasing the variability at this particular stage. An explanation of
257 this phenomenon is attempted in the following paragraphs, by analysing the single PNC evolutions
258 and the results from the second field campaign.

259 Despite the variability described above, some common features can indeed be found in the
260 evolution of the PNCs measured in these various test cases. Fig. 4 reports a selection from the total
261 available 65 cases illustrating some of the most common patterns during the ground-fixed

measurements. For instance, the pattern visualised in Fig. 4a is one of the most common (around 28% of the runs), even though the numerical values can differ from case to case. This pattern is very similar to the one presented in a preliminary field campaign by Kumar et al. (2009c). It features a rapid building of PNC, with a peak usually before 1 s (see Table 1), then a rapid decrease followed by a slightly slower decay before returning to the background values. A slightly modified version of this pattern is presented in Fig. 4b (another 15% of the runs share this trend). This latter situation is more common for higher car velocities where, as also shown in Table 1, the decay time is longer.

Fig. 4c shows another common pattern (around 32% of the runs) where a larger peak followed by another smaller peak can be observed. This feature was frequently observed in experimental cases with higher car speeds at 0.25 m height. This pattern could probably be caused by wake effects i.e. increasing turbulence with car speeds (see, e.g., Di Sabatino et al., 2003). The pattern in Fig. 4d shows another frequently seen pattern (the remaining 25%) where a smaller peak rapidly follows a larger peak. This pattern was often observed during the measurements at $H = 0.25$ m, especially at lower vehicle speeds, and lead to the average evolution profile highlighted in Fig. 3b and 3d ($H = 0.25$ m, $V = 20$ and 30 km h⁻¹). This pattern could also be due to rapid movements of the vehicle wake; we will come back on this point with more details in the next section (3.2.2), where on-board measurements are analysed. PNC evolutions for the entire dataset are available on request.

The evolution of PNCs measured at a ground-fixed sampling point is mainly driven by nucleation and dilution. As a matter of facts, the effect of the emission measured at the fixed location lasts only for a few seconds (usually about 5–15 s, see also Table 1). Within this time scale, coagulation and deposition are not likely to play a significant role in PNC evolution (see Zhang and Wexler, 2004, Ketzel and Berkowicz, 2004, and the review by Carpentieri et al., 2011). Other transformation processes such as condensation do not alter the PNCs (Kumar et al., 2010). Possible effects of various transformation processes on number and size distributions in the studied cases are further analysed in detail in Section 3.3.

3.2.2 PNCs during on-board measurements

288 A summary of the results for the on-board measurements is presented in Tables 2, 3 and 4
289 for vehicle speed equal to 20, 30 and 40 km h⁻¹, respectively. Data in the tables represents averages
290 over all the runs in same nominal conditions (i.e. car speed and measurement point) as described in
291 Section 2.2 and Fig. 1.

292 Results presented in Figs. 5 and 6 highlight the spatial distribution of total PNCs in the car wake. As
293 expected, the maximum PNCs are measured along the points in line with the tailpipe. The closest
294 measurement points are P_a and P_d (Fig. 5a); these are respectively at $z=0.10$ and 0.50 m above the
295 ground and both at $x=0.45$ m away from the tailpipe. However, PNCs measured at P_a are 2.5–2.7
296 times higher than those found at P_d in all the experimental cases. This could be explained
297 considering the fact that P_a is within the direct emission plume from the tailpipe, while P_d is in the
298 recirculation wake.

299 An interesting aspect of the on-board measurements is the large differences in PNC among the 3
300 measurement points at $z=0.10$ m (Fig. 5) for all car speeds. PNCs are much more uniform at $z=0.50$
301 m and $z=0.90$ m, where the plume is within the recirculation zone (Hucho, 1987) rather than
302 entrained in the air flow coming from below the car. This huge difference between PNCs at $y=-0.45$
303 m (in line with the tailpipe) and PNCs at other positions along the y -axis can partially explain the
304 large variability observed during the ground-fixed measurements (Section 3.2.1). Even small
305 deviations from the planned lateral position of the passing car could have generated large
306 differences in the measured concentrations, if the sampling point was only a few centimetres further
307 away from the tailpipe (see Section 3.4).

308 The above observations also provide a possible explanation for the results presented in Section 3.2.1
309 about the ground-fixed measurements at $z=0.25$ m. This height is very close to the bottom of the
310 car, where the interface between the two behaviours observed above (the uniform PNCs observed
311 within the wake and the PNCs measured in the direct flow below the car) is probably located (see
312 Fig. 7). The double peaks often observed at $z=0.25$ m during the ground-fixed measurements could

313 be due to the fact that the sampling point was moving in and out from the wake (or, to be more
314 precise, the wake moved, while the ground-fixed point was, indeed, fixed). No wonder the
315 variability observed during the $z=0.25$ m cases is higher than that observed at $z=0.10$ m.

316 At the same measurement points, the PNCs measured at higher car speeds (30 and, especially, 40
317 km h^{-1}) are generally up to a factor of 3 larger than those measured at 20 km h^{-1} . As for the analysis
318 of ground-fixed measurements (Section 3.1.1), this could be due to a combination of two factors: (i)
319 particle number emissions at higher speed are greater (Kumar et al., 2011b), and (ii) measurement
320 points at the same distance from the tailpipe are actually closer, from a temporal point of view, at
321 higher speeds.

322 **3.3. Transformation of number and size distributions**

323 **3.3.1. PNDs during ground-fixed measurements**

324 The analysis presented in this paragraph included the study of PNDs and GMDs at various
325 stages during the measurements. PNDs, in particular, are presented in terms of normalised number
326 weighted size spectrum. This normalisation has been carried out by dividing the measured number
327 spectral densities ($dN/d\log D_p$) for each size category (characterised by the diameter D_p) by the total
328 PNC at that particular time. This kind of normalisation was done for highlighting the relative
329 transformations in terms of size spectrum, removing the effect of dilution. In facts, dilution affects
330 all particle size ranges uniformly, and plotting the normalised spectra at different time steps on the
331 same graph will show a perfect superposition of those spectra when dilution is the only process
332 involved.

333 As in the case of PNCs, common features of the evolution of PNDs can be observed across the
334 various experimental runs, despite the expected differences among different runs even in nominally
335 identical conditions. Fig. 8 shows a typical behaviour of the normalised size spectrum during the
336 three main stages (pre-evolution, evolution and post-evolution, as defined below). The subdivision
337 of the transformation process in different stages is somewhat arbitrary, and it was done by observing
338 common trends of the PNDs during the measurements. Up to four evolution sub-stages, each

339 showing a distinct PND evolution pattern during various experimental runs, were also identified.

340 The “pre–evolution” stage represents the status of the size spectrum just before $t=0$ s (as defined in
341 Section 3.2), i.e. site background conditions. During this stage the PNDs are, as expected, consistent
342 across all the experimental cases, except for few observed differences due to uncontrolled external
343 disturbances. The peak number spectral densities during this stage are usually in the 10–12 nm size
344 range and the PNDs are dominated mainly by nucleation mode particles (Fig. 8a). “Evolution” stage
345 is defined as the stage where most of the transformation processes occur; this is assumed to start at
346 $t=0$ s. In the case exemplified in Fig. 8, and common to most of the runs in the first experimental
347 case (i.e. $H=0.10$ m, $V=20$ km h⁻¹), the evolution stage can be further divided in two sub–stages,
348 marking a different behaviour in the transformation process: during the first sub–stage (tagged
349 “evolution 1” in Fig. 8), there is a rapid change in the normalised size spectrum. This shows an
350 initial increase in spectral densities at diameters around 25 nm followed by an even greater increase
351 for particle sizes between ~40 and ~100 nm (Fig. 8b). The second sub–stage of evolution (tagged
352 “evolution 2”) starts at $t = 0.8$ s and shows the normalised size spectrum returning back to the site
353 background, very similar to the PNDs observed in the pre–evolution stage (Fig. 8c). From $t=1.9$ s
354 onwards (Fig. 8d), the normalised spectrum does not deviate much from the site background and
355 only total PNC variations are observed (dilution–only; see also Fig. 9a). This stage is defined as
356 “post–evolution”.

357 Fig. 9a shows the evolution of the GMDs, and the corresponding PNCs, during all the stages
358 highlighted in Fig. 8. GMDs increase rapidly from the background value ~15 nm to a peak of ~38
359 nm during the ‘evolution 1’ sub–stage due to the freshly–emitted particles in the nucleation mode
360 from our car (Fig. 8), followed, with the same rapidity, by a decrease to the background value in
361 ‘evolution 2’ sub–stage. Comparing the GMD trend with the corresponding PNCs (Fig. 9a), it can
362 be noted that the former is slightly quicker in its return to the background conditions. This means
363 that at the beginning of the post–evolution stage we still have a PNC variation, despite the fact that
364 the GMD and the normalised spectrum do not change much (dilution only). This is, however, not

365 always the case, as discussed below.

366 The pattern outlined above, with two evolution sub-stages, is also common to the second
367 experimental set ($H=0.10$ m, $V=30$ km h⁻¹), although in this case another behaviour can also be
368 observed (see Fig. 10): besides both the evolution sub-stages seen in earlier cases ('evolution 1' and
369 'evolution 3' in Fig. 10, corresponding to evolution 1 and 2 in Fig. 8), another sub-stage (evolution
370 2' in Fig. 10) is present. During this additional sub-stage size distributions do not change in relative
371 terms; the remarkable superposition of the various normalised spectra suggests dilution as the only
372 acting process. This sub-stage usually started after about 1 s, and lasted for another 2 or 3 s (see
373 Fig. 10c), followed by a very slow return (Fig. 10d) to the original distribution (Fig. 10e). This trend
374 is exemplified in Fig. 9b, for the same experimental case, with the help of GMD and PNC
375 evolutions. While the PNC evolution curve is very similar to that in Fig. 9a, the GMD pattern is
376 utterly different, with a fast initial increase up to about 24 nm and then a slow return to the initial
377 value (~15 nm). In this second phase, dilution is usually the main transformation process. This
378 behaviour can also be observed frequently in other two experimental cases (i.e. $H = 0.10$ m, $V = 40$
379 and 50 km h⁻¹).

380 Similar patterns can be identified in other four experimental cases at $H = 0.25$ m ($V = 20, 30, 40$ and
381 50 km/h), except an additional fourth evolution sub-stage during the initiation of evolution process
382 is often observed (see Fig. 11). The evolution sub-stages 2, 3 and 4 in Fig. 11c, d and e,
383 respectively, look identical to those found in evolution sub-stages 1, 2 and 3 in Fig. 10b, 10c and
384 10d. The additional sub-stage (Fig. 11b) at the beginning of the evolution process in the present
385 case shows an initial rise in number spectral densities around the 17 nm diameter, followed by
386 another increase in the 23–30 nm range and then a rapid decrease. After about 1 s from $t=0$, this
387 trend is generally over, and the usual evolution pattern already observed in Fig. 10 is established
388 again. This process can also be observed in the evolutions of the GMDs and PNCs (Fig. 9c), and
389 can be explained by the fact that different parts of the emitted plume are being sampled at different
390 times, as analysed during the on-board measurements and further discussed in Section 3.4.

391 3.3.2 PNDs during on-board measurements

392 Fig. 12 shows the particle size spectra during the on-board measurements at P_a ($x=0.45$ m,
393 $y=-0.45$ m, $z=0.10$ m) and P_d ($x=0.45$ m, $y=-0.45$ m, $z=0.50$ m) at different car speeds; both these
394 points are in line with the tailpipe as illustrated in Figs. 1, 5a and 6a.

395 Apart from the numerical value of PNC in different runs, which is likely to be sensitive to both the
396 mechanical (traffic) and atmospheric (wind) produced turbulence (Kumar et al., 2008a), the shapes
397 of the PNDs for different runs within the same experimental condition (car speed and measurement
398 point) are reasonably similar. The magnitude of accumulation mode particles seem to be more
399 affected by the above flow effects than the nucleation mode particles in different runs of the same
400 experimental cases. PNDs at P_a (Figs. 12a, b and c) appear to be more affected by the different car
401 speed (or rather, as pointed out in Section 3.2.2, by the distance from the tailpipe, from a time scale
402 point of view) than PNDs at P_d (Figs. 12d, e and f), which are similar to each other. In particular, the
403 lowest measurement point is unswervingly affected by particles (mainly in the 30–45 nm size range
404 as evident from a distinct peak at P_a ; Figs. 12a, b and c) coming directly from the tailpipe; this peak
405 in the PNDs is nearly absent at $z=0.50$ m (P_d ; Figs. 12d, e and f). Most of the measured particles at
406 both measurement points (i.e. P_a and P_d) are in the range 60–160 nm, as also evident in Tables 2, 3
407 and 4 from PNC–1, PNC–2, GMD–tot, GMD–1 and GMD–2 values. The bimodal shape of the
408 PNDs is more evident at P_a for higher car speeds. This is due to the fact that freshly emitted fine
409 particles are sampled earlier at 40 km h^{-1} than at 20 km h^{-1} .

410 PNDs at P_g ($x=0.45$ m, $y=-0.45$ m, $z=0.90$ m) and P_j ($x=0.80$ m, $y=-0.45$ m, $z=0.50$ m) are very
411 similar to those observed at P_d (Figs. 12d, e and f), and are not presented here for brevity reasons;
412 however, their results are summarised in Tables 2, 3 and 4. Numerically, the differences between the
413 average PNDs at these measurement points (P_d , P_g and P_j) are very small in all experimental
414 conditions. As shown in Tables 2, 3 and 4, GMDs at points P_d and P_g , in particular, seem not to be
415 greatly affected by the car speed. On the contrary, at points P_a and P_j , car velocity seems to have a
416 slightly greater influence, driven mostly by accumulation mode particles, while GMDs and PNCs in

417 the range 5–30 nm do not show much sensitivity to this parameter.

418 Figs. 13a, b and c show PNDs at P_b ($x=0.45$ m, $y=0$, $z=0.10$ m; car centre line). The strong influence
419 of smaller particles on the total PNDs is clearly visible in these figures. Further evidence of this
420 influence is given by the total GMD values (Tables 2, 3 and 4) which are sensibly lower than the
421 other measurement points in all the experimental conditions. Another peculiarity of this
422 measurement point is the size distribution of nucleation mode particles: the GMD for particles <30
423 nm is in the range 10–13 nm, while for most of the other measurement points this value is usually
424 between 16 and 21 nm. These low values can only be found at one other measurement point,
425 notably at P_c ($x=0.45$ m, $y=0.45$ m, $z=0.10$ m).

426 Spectra measured at other centre line points (specifically P_e , $x=0.45$ m, $y=0$, $z=0.50$ m, and P_h ,
427 $x=0.45$ m, $y=0$, $z=0.90$ m) are similar in shape to those at P_d and P_g . On the contrary, small
428 differences can be observed at P_k ($x=0.80$ m, $y=0$, $z=0.50$ m, see Figs. 13d, e and f), with a slightly
429 higher spectral density in the 20–40 nm size range, and a maximum spectral density value at smaller
430 diameters (50–85 nm), tending to even decrease as the car speed increases. This is also reflected in
431 total GMDs values (Tables 2, 3 and 4) which become smaller as the speed increases.

432 PNDs measured at P_c ($x=0.45$ m, $y=-0.45$, $z=0.10$ m) are presented in Figs. 14a, b and c for
433 different car speeds. These figures share a few similarities with P_b , and this is also evidenced by the
434 data in Tables 2, 3 and 4, as already pointed out. These points seem to be at the interface between
435 two different zones (the recirculation wake and the zone where the flow is coming from below the
436 car, see Fig. 7 and Section 3.2.2) of the wake, and this is reflected in the variability observed during
437 different runs at nominally the same experimental conditions. Other similarities can be observed
438 between PNDs at P_i ($x=0.45$ m, $y=0.45$, $z=0.90$ m; Figs. 14d, e and f) and at P_k (Figs. 13d, e and f).
439 PNDs at the other remaining measurement points (P_f , $x=0.45$ m, $y=0.45$ m, $z=0.50$ m, and P_l , $x=0.80$
440 m, $y=0.45$ m, $z=0.50$ m) have the common shape already seen for P_d , P_e , P_g , P_j and P_h .

441 3.4. Discussion

442 The large amount of data gathered during the two experimental campaigns offer several
443 points for discussion. The adopted experimental strategy allowed us to look at different aspects of
444 the nanoparticle dispersion process from a moving car, integrating the results from previous studies,
445 performed with different measurement setups. Particularly, the present study took into account the
446 effects of the vehicle wake on the nanoparticle transformation process, thanks to both the fast
447 response instrument and the integrated experimental methodology used.

448 Some interesting observations can be made about the various transformation processes involved in
449 the first stages from the emission of nanoparticles from the car tailpipe. Carpentieri et al. (2011)
450 reported in their review that particles emitted by cars are subject to a number of transformation
451 processes (i.e. nucleation, dilution, coagulation, condensation and deposition) which are discussed
452 below in the context of our results.

453 Dilution is the main process acting on nanoparticles emitted by diesel cars (Zhang and Wexler,
454 2004; Ketzel and Berkowicz, 2004; Kumar et al., 2009b). It is mainly driven by the vehicle–
455 produced turbulence in the near wake region while the atmospheric turbulence dominates this in far
456 wake regions (Carpentieri et al., 2011; Kumar et al., 2011b). As soon as the exhaust emissions are
457 released from the tailpipe, they start to dilute at a rapid rate which causes sulphuric acid–induced
458 nucleation (Zhang and Wexler, 2004). The interaction between nucleation and dilution is very fast,
459 and it has been rarely captured by experimental measurements under actual driving conditions.
460 Across all our ground–fixed measurements, this effect can be clearly observed by looking at the fast
461 growing PNCs in nucleation mode in the very first stage of evolution (see Figs. 8b, 10b, 11b and
462 11c). The process is usually over within 1 s, confirming the preliminary results by Kumar et al.
463 (2009c) as well as the study conducted by Rönkkö et al. (2006) and (2007). The latter two studies
464 conducted both on–road (with the chasing method) and in laboratory measurements of the number
465 and size distributions of the particles. They found that fast dynamical processes (mainly nucleation)
466 in the exhaust plume of a truck and a bus, respectively, had already taken place within 0.7 s from
467 emission, confirming our current observations.

468 Irrespective of vehicle speed, results from the ground–fixed measurements show that the PNCs
469 reached to a peak after a remarkably constant time. This aspect indicates a negligible influence of
470 the vehicle wake in the very first phases of evolution, where dilution and nucleation are likely to be
471 driven mainly by the immediate interaction between the plume jet and the external environment.

472 After reaching the PNC peak, the nanoparticles start mixing with the wake or the external flow
473 (depending on which part of the plume we are looking at). This is evidenced by the longer decay
474 time experienced by PNC measurements in the ground–fixed experiments with higher car speed (for
475 example, see Table 1). PNDs measured in this phase of the evolution, show another transformation
476 process acting on the emitted nanoparticles, besides dilution. A growth in accumulation mode
477 particles, in particular, is driven by condensation of super-saturated vapours (Zhang and Wexler,
478 2004). The time scale associated with this process varies from case to case. However, it seems to
479 last for few seconds, after which the measured plume becomes indistinguishable from the
480 background; this finding is also in agreement with preliminary results by Kumar et al. (2009c).

481 Effects of dry deposition and coagulation on PNCs cannot easily be captured, even indirectly, by
482 our experimental setups. According to previous studies (Zhang and Wexler, 2004; Ketzel and
483 Berkowicz, 2004) the time scales associated with these transformation processes are comparable, or
484 even higher, than the duration of our measurements. Thus, when the particles are released from the
485 tailpipe, coagulation and deposition could start playing a significant role only after a few seconds,
486 and certainly many metres away from the car. However, some of the emitted nanoparticles are
487 certainly entrained within the vehicle recirculation wake and this increases the residence time
488 making it possible for these two processes to affect PNCs and PNDs.

489 In fact, on–board measurements at different locations behind the moving car, as well as ground–
490 fixed measurements at different heights above the road level, showed different behaviours for
491 nanoparticles sampled within the vehicle wake and those sampled, supposedly, in the flow outside
492 its recirculation vortices. PNDs measured along the direct emission line from the tailpipe show a

493 typical bimodal shape when very close to the release point, while a prevalence of accumulation
494 mode particles can be observed further away. On the contrary, all samples taken within the vehicle
495 wake, even relatively near the tailpipe, displayed the common feature of PNDs dominated by
496 accumulation mode particles. This is further evidence that nanoparticles in the vehicle wake reside
497 for a longer time, showing characteristics radically different from new, freshly emitted, particles.
498 These results are comparable to those reported by Wehner et al. (2009) and Morawska et al. (2007).
499 In the former study, the sampling point was fixed on the z and y axes, with two locations at $x=0.45$
500 m and $x=0.90$ m from a diesel car; their measurements at $x=0.90$ m are very similar to our
501 measurements at $x=0.80$ m, while results at $x=0.45$ m show different behaviour in different
502 experimental conditions; the sampling point in that case is probably at the border between the
503 recirculation wake and external flow, and this could partially explain the fact that nucleation mode
504 particles were not always detected by the authors of that study. In the second work (Morawska et
505 al., 2007) the sampling point was both within the wake and far from the tailpipe, so that nucleation
506 mode particles could not be observed.

507 The above finding about the differences between particles sampled within the recirculation wake,
508 and those sampled within the external flow is also well in agreement with the numerical simulations
509 carried out by Chan et al. (2010) for an exhaust plume in a vehicle wake, and has at least two strong
510 implications. Firstly, mathematical models dealing with nanoparticle dispersion at this local scale
511 should take particles recirculated within, and later escaping from, the wake into account, bearing in
512 mind that their characteristics are different from those freshly emitted. Further information on wake
513 residence time, as well as an estimate on the influence of coagulation and dry deposition processes
514 will be needed for a proper characterisation of the particles escaping from the wake.

515 The second implication is about the measurement methodologies generally used for this kind of
516 studies. As briefly reviewed in Section 1, essentially three different methodologies have been used
517 for measuring nanoparticles emitted from moving vehicles at close distance from the emission
518 point. The chasing method, as already discussed in the introduction, cannot be reliably used to

519 characterise nanoparticles in the very first stages from emission, unless by using trailing vehicles
520 which heavily modify the natural flow behind the vehicle. On-board measurements, as
521 demonstrated in this work and by Wehner et al. (2009), can be very effective in this kind of studies.
522 Our results, however, show that careful positioning of the sampling points is needed to properly
523 characterise the dispersion and transformation of nanoparticles. In particular, the measurement grid
524 must be set up according to which type of nanoparticles the study is trying to characterise: new
525 freshly emitted particles, relatively aged wake-recirculated particles or both; especially in the latter
526 case, either a detailed knowledge of the vehicle wake or a large (and high resolution) grid is needed.

527 Ground-fixed measurements can give us continuous information on nanoparticle transformations
528 both in space and time; however, they must be carefully interpreted. Given the uncertainties on the
529 lateral positioning of the passing vehicle, it is not easy to control the experimental conditions. As
530 exemplified in Fig. 15, given the shape of the direct plume emitted from the tailpipe, even small
531 deviations from the plume centreline, say a few centimetres, can cause very different parts of the
532 plume to be sampled during different runs. With the usual experimental setup, there is no easy way
533 to establish the distance from the tailpipe at $t=0$. Furthermore, when sampling the plume at greater
534 distances from the tailpipe, or higher levels from the ground, the influence of the wake-recirculated
535 plume will not be negligible.

536 **4. Summary and conclusions**

537 Two sets of experimental campaigns were carried out using a fast response particle
538 spectrometer to measure number and size distributions of nanoparticles in the wake of a moving
539 diesel car. Both ground-fixed and on-board setups were used. This experimental strategy allowed
540 us to take into account the effect of the vehicle wake on the nanoparticle dispersion process,
541 providing also new keys for the interpretation of results from previous studies by other authors.

542 The measurements again confirmed the dilution as a main driver for controlling the transformation
543 of nanoparticles in the emitted plume of a diesel car. Results about the first evolution phase,

544 immediately after emission, agreed with previous literature and confirmed the presence of a first
545 phase in which PNCs and PNDs change rapidly due to nucleation processes driven by quickly
546 increasing dilution. This stage is over after a few tenths of second, when condensation processes
547 start to become effective in increasing the size of the particles, with a fast (but slower than the first
548 phase) increase of accumulation mode particles.

549 What happens next is less clear due to complex interactions between the flow in vehicle wake and
550 exhaust emissions that start to affect the dispersion and transformation of nanoparticles. The
551 combined analysis of ground-fixed measurements, on-board measurements and past literature
552 clearly showed the presence of two different groups of nanoparticles in the volume immediately
553 adjacent to the back of the moving vehicle: the new particles freshly emitted from the tailpipe, and
554 the relatively aged particles in the flow recirculation wake of the vehicle. The two groups have
555 experienced different transformation processes with different time scales, strongly linked with the
556 dilution levels they are subject to. While in the immediate distance in front of the tailpipe the
557 freshly emitted group of new nanoparticles is abundantly dominant, in most of the volume above
558 the tailpipe level and close to the back of the car only the relatively aged group can be found. Other
559 zones, further away from the vehicle, contain a mixture of both, and particular care should be taken
560 in interpreting the results from measurement campaigns. This aspect related to the vehicle wake is
561 often overlooked in past studies.

562 The DMS50, used in this study to measure nanoparticle number concentrations, could be employed
563 in future works in conjunction with additional instruments to attempt an estimation of dry
564 deposition, e.g. by using an ultrasonic anemometer to apply the eddy covariance method (Buzorius
565 et al., 1998; Buzorius et al., 2003), or the nanoparticle emission factor using a tracer gas
566 measurement system (Hak et al., 2009; Wehner et al., 2009). A detailed knowledge of the flow and
567 dispersion characteristics of the vehicle wake could also further help interpreting the large amount
568 of data gathered during this experimental campaign. Our next step is to carry out a detailed
569 assessment of the wake behind the experimental car by means of wind tunnel tests in controlled

570 conditions on reduced scale models of our experimental car. This experimental study will allow us
571 to characterise the dilution process in the vehicle wake by combining high resolution concentration
572 measurements of a passive tracer gas and three dimensional flow velocity components, and their
573 comparison with the nanoparticle measurements presented here. These will also help in analysing
574 the competing influences and interactions of various transformation processes with each other.

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583 **6. References**

- 584 Buzorius, G., Rannik, U., Makela, J.M., Vesala, T., Kulmala, M., 1998. Vertical aerosol particle
585 fluxes measured by eddy covariance technique using condensational particle counter. *Journal*
586 *of Aerosol Science* 29, 157-171.
- 587 Buzorius, G., Rannik, Ü., Nilsson, E.D., Vesala, T., Kulmala, M., 2003. Analysis of measurement
588 techniques to determine dry deposition velocities of aerosol particles with diameters less than
589 100 nm. *Journal of Aerosol Science* 34, 747-764.
- 590 Carpentieri, M., Kumar, P., Robins, A., 2011. An overview of experimental results and dispersion
591 modelling of nanoparticles in the wake of moving vehicles. *Environmental Pollution* 159,
592 685-693.
- 593 Chan, T.L., Liu, Y.H., Chan, C.K., 2010. Direct quadrature method of moments for the exhaust
594 particle formation and evolution in the wake of the studied ground vehicle. *Journal of Aerosol*
595 *Science* 41, 553-568.
- 596 Di Sabatino, S., Kastner-Klein, P., Berkowicz, R., Britter, R.E., Fedorovich, E., 2003. The
597 Modelling of Turbulence from Traffic in Urban Dispersion Models – Part I: Theoretical
598 Considerations. *Environmental Fluid Mechanics* 3, 129-143.
- 599 Giechaskiel, B., Nziachristos, L., Samaras, Z., Scheer, V., Casati, R., Vogt, R., 2005. Formation

600 potential of vehicle exhaust nucleation mode particles on-road and in the laboratory.
 601 Atmospheric Environment, 39, 3191-3198.

602 Hak, C.S., Hallquist, M., Ljungstrom, E., Svane, M., Pettersson, J.B.C., 2009. A new approach to
 603 in-situ determination of roadside particle emission factors of individual vehicles under
 604 conventional driving conditions. Atmospheric Environment 43, 2481-2488.

605 Hinds, W.C., 1999. Aerosol Technology: Properties, Behaviour and Measurement of Airborne
 606 Particles. John Wiley & Sons, UK.

607 Hucho, W.-H., 1987. Aerodynamics of road vehicles. SAE Technical paper ISBN, 0-7680-0029-7.

608 Ketzel, M., Berkowicz, R., 2004. Modelling the fate of ultrafine particles from exhaust pipe to rural
 609 background: an analysis of time scales for dilution, coagulation and deposition. Atmospheric
 610 Environment 38, 2639-2652.

611 Kittelson, D.B., Watts, W.F., Johnson, J.P., 2006. On-road and laboratory evaluation of combustion
 612 aerosols - Part 1 : Summary of deisel engine results. Journal of Aerosol Science 37, 913-930.

613 Kumar, P., Fennell, P., Britter, R., 2008a. Effect of wind direction and speed on the dispersion of
 614 nucleation and accumulation mode particles in an urban street canyon. Science of the Total
 615 Environment 402, 82-94.

616 Kumar, P., Fennell, P., Britter, R., 2008b. Measurements of particles in the 5-1000 nm range close to
 617 road level in an urban street canyon. Science of the Total Environment 390, 437-447.

618 Kumar, P., Fennell, P., Symonds, J., Britter, R., 2008c. Treatment of losses of ultrafine particles in
 619 long sampling tubes during ambient measurements. Atmospheric Environment 42, 8819-8826.

620 Kumar, P., Fennell, P., Hayhurst, A., Britter, R.E., 2009a. Street versus rooftop level concentrations
 621 of fine particles in a Cambridge street canyon. Boundary-Layer Meteorology 131, 3-18.

622 Kumar, P., Garmory, A., Ketzel, M., Berkowicz, R., Britter, R., 2009b. Comparative study of
 623 measured and modelled number concentrations of nanoparticles in an urban street canyon.
 624 Atmospheric Environment 43, 949-958.

625 Kumar, P., Robins, A., Britter, R., 2009c. Fast response measurements for the dispersion of
 626 nanoparticles in a vehicle wake and in a street canyon. Atmospheric Environment 43, 6110-
 627 6118.

628 Kumar, P., Robins, A., Vardoulakis, S., Britter, R., 2010. A review of the characteristics of
 629 nanoparticles in the urban atmosphere and the prospects for developing regulatory control.
 630 Atmospheric Environment 44, 5035-5052.

631 Kumar, P., Gurjar, B.R., Nagpure, A., Harrison, R.M., 2011a. Preliminary estimates of nanoparticle
 632 number emissions from road vehicles in megacity Delhi and associated health impacts.
 633 Environmental Science and Technology, in press, doi: 10.1021/es2003183.

634 Kumar, P., Ketzel, M., Vardoulakis, S., Pirjola, L., Britter, R., 2011b. Dynamics and dispersion
 635 modelling of nanoparticles from road traffic in the urban atmospheric environment – a review.

636 Journal of Aerosol Science, in press.

637 Morawska, L., Ristovski, Z.D., Johnson, G.R., Jayaratne, E.R., Mengersen, K., 2007. Novel method
 638 for on-road emission factor measurements using a plume capture trailer. Environmental
 639 Science and Technology 41, 574-579.

640 Rönkkö, T., Virtanen, A., Kannosto, J., Keskinen, J., Lappi, M., Pirjola, L., 2007. Characteristics of
 641 nucleation mode particles in exhaust of Euro IV heavy duty diesel vehicle. Environmental
 642 Science and Technology 41, 6384-6389.

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

646 Vogt, R., Scheer, V., Casati, R., Benter, T., 2003. On-road measurement of particle emission in the
 647 exhaust plume of a diesel passenger car. Environmental Science and Technology 37, 4070-
 648 4076.

649 Wehner, B., Uhrner, U., von Löwis, S., Zallinger, M., Wiedensohler, A., 2009. Aerosol number size
 650 distributions within the exhaust plume of a diesel and a gasoline passenger car under on-road
 651 conditions and determination of emission factors. Atmospheric Environment 43, 1235-1245.

652 Zhang, K.M., Wexler, A.S., 2004. Evolution of particle number distribution near roadways - Part I:
 653 analysis of aerosol dynamics and its implications for engine emission measurement.
 654 Atmospheric Environment 38, 6643-6653.

655

656 **Figure captions**

657 Fig. 1. Schematic of the set up for both the ground-fixed (Site 1) and on-board (Site 2)
658 measurement campaigns (not in scale).

659 Fig. 2. Comparison of the noise level of the DMS50 at different sampling frequencies (0.1, 1 and
660 10 Hz) with the typical background PNDs, representing the minimum concentrations to be
661 measured at the sampling site. Error bars represent standard deviation of 20 PND samples taken at
662 10 Hz sampling frequency.

663 Fig. 3. Temporal evolution of normalised PNCs for ground-fixed measurements, averaged over the
664 valid runs for each experimental case; error bars represent the standard errors.

665 Fig. 4. Figures showing typically observed temporal evolution patterns of PNCs for ground-fixed
666 measurements, experimental cases (a): $H = 0.10$ m, $V = 20$ km h⁻¹, Run 02, (b) $H = 0.10$ m, $V = 40$
667 km h⁻¹, Run 07, (c) $H = 0.25$ m, $V = 40$ km h⁻¹, Run 04, and (d) $H = 0.25$ m, $V = 20$ km h⁻¹, Run 08.

668 Fig. 5. PNCs (# cm⁻³) measured at $x=0.45$ m during the on-board experiments: (a) map, (b) $V=20$
669 km h⁻¹, (c) $V=30$ km h⁻¹, and (d) $V=40$ km h⁻¹. Area of each bubble corresponds to magnitude of
670 PNCs at that point.

671 Fig. 6. PNCs (# cm⁻³) measured at $z=0.50$ m during the on-board experiments: (a) map, (b) $V=20$
672 km h⁻¹, (c) $V=30$ km h⁻¹, and (d) $V=40$ km h⁻¹. Area of each bubble corresponds to magnitude of
673 PNCs at that point.

674 Fig. 7. Simplified representation of the interaction between the recirculation wake and the sampling
675 points for both ground-fixed and on-board measurements. Note that wake position and extension
676 can vary both in time and space due to turbulence and instabilities.

677 Fig. 8. Normalised size distributions during (a) pre-evolution, (b) evolution 1, (c) evolution 2, and
678 (d) post-evolution for the experimental case: $H = 0.10$ m, $V = 20$ km h⁻¹, Run 01.

679 Fig. 9. Variation of GMDs and normalised PNCs (PNCs divided by the corresponding peak values)
680 with time in various transformation stages for experimental cases: (a) $H = 0.10$ m, $V = 20$ km h⁻¹,
681 Run 01, (b) $H = 0.10$ m, $V = 30$ km h⁻¹, Run 06, (c) $H = 0.25$ m, $V = 20$ km h⁻¹, Run 08.

682 Fig. 10. Normalised size spectra during (a) pre-evolution, (b) evolution 1, (c) evolution 2, (d)
683 evolution 3 and (e) post-evolution for the experimental case: $H = 0.10$ m, $V = 30$ km h⁻¹, Run 06.

684 Fig. 11. Normalised size spectra during (a) pre-evolution, (b) evolution 1, (c) evolution 2, (d)
685 evolution 3, (e) evolution 4 and (f) post-evolution for the experimental case: $H=0.25$ m, $V=20$ km h⁻¹,
686 Run 08.

687 Fig. 12. Particle size distributions for on-board measurements at P_a for (a) $V=20$ km h⁻¹, (b) $V=30$
688 km h⁻¹, (c) $V=40$ km h⁻¹, and P_d for (d) $V=20$ km h⁻¹, (e) $V=30$ km h⁻¹, (f) $V=40$ km h⁻¹.

689 Fig. 13. Particle size distributions for on-board measurements at P_b for (a) $V=20$ km h⁻¹, (b) $V=30$
690 km h⁻¹, (c) $V=40$ km h⁻¹, and at P_k for (d) $V=20$ km h⁻¹, (e) $V=30$ km h⁻¹, (f) $V=40$ km h⁻¹.

691 Fig. 14. Particle size distributions for on-board measurements at P_c for (a) $V=20$ km h⁻¹, (b) $V=30$
692 km h⁻¹, (c) $V=40$ km h⁻¹, and at P_i for (d) $V=20$ km h⁻¹, (e) $V=30$ km h⁻¹, (f) $V=40$ km h⁻¹.

693 Fig. 15. Simplified representation of the effects of the lateral positioning of the ground-fixed
694 sampling on the detected plume at two different hypothetical runs (Run1 and Run2); note that the
695 wake-recirculated plume has not been considered in this representation.

696 **List of Tables**

697 Table 1. Summary of the results from the ground-fixed measurements; values expressed are in
698 terms of mean $\pm$ standard deviation.

Experimental case	Valid runs	Sampling height (H) and car speed (V)		Average peak PNC ($\times 10^5 \# \text{ cm}^{-3}$)	Average time (s) at peak PNCs	Average duration (s) of event
		H (m)	V (km h $^{-1}$)			
1	8	0.10	20	5.19 ± 3.33	0.8 ± 0.2	5.6 ± 3.6
2	9	0.10	30	9.20 ± 6.68	0.8 ± 0.2	6.3 ± 2.7
3	9	0.10	40	2.60 ± 2.41	0.8 ± 0.2	7.9 ± 3.6
4	6	0.10	50	6.58 ± 5.81	1.6 ± 1.2	9.1 ± 6.2
5	10	0.25	20	3.39 ± 3.65	1.8 ± 0.4	6.1 ± 4.6
6	10	0.25	30	6.73 ± 5.62	1.7 ± 0.9	6.7 ± 3.8
7	9	0.25	40	5.20 ± 4.20	1.2 ± 0.2	5.5 ± 1.8
8	4	0.25	50	1.95 ± 1.55	1.3 ± 0.4	10.6 ± 4.1

699

700 Table 2. Summary of the results from the on-board measurements with $V=20 \text{ km h}^{-1}$. Here and in
701 the tables hereafter PNC-1 and GMD-1 values refer to particles with $D_p < 30 \text{ nm}$ (nucleation
702 mode), while PNC-2 and GMD-2 refer to particles with $30 < D_p < 300$ (accumulation mode).GMD
703 stands for geometric mean diameter.

704

Measurement points				Total PNC ($\times 10^5 \# \text{ cm}^{-3}$)	Total GMD (nm)	PNC-1 ($\times 10^5 \# \text{ cm}^{-3}$)	GMD-1 (nm)	PNC-2 ($\times 10^5 \# \text{ cm}^{-3}$)	GMD-2 (nm)	Valid Runs
ID	x (cm)	y (cm)	z (cm)							
P _a	45	-45	10	43.04	61.89	5.69	19.26	37.35	74.14	4
P _b	45	0	10	2.23	46.77	0.58	12.76	1.65	71.09	3
P _c	45	45	10	0.69	88.43	0.64	13.39	0.63	111.21	5
P _d	45	-45	50	16.35	62.20	1.43	19.81	14.93	69.72	2
P _e	45	0	50	7.31	65.69	0.71	16.79	6.59	75.44	2
P _f	45	45	50	3.25	65.17	0.30	16.54	2.94	77.03	2
P _g	45	-45	90	4.15	69.72	0.32	20.64	3.83	77.69	2
P _h	45	0	90	4.14	74.48	0.22	21.71	3.93	80.03	2
P _i	45	45	90	2.15	67.23	0.26	18.22	1.89	81.66	2
P _j	80	-45	50	8.76	74.01	0.54	20.88	8.21	80.83	2
P _k	80	0	50	5.38	70.77	0.47	17.50	4.91	80.71	2
P _l	80	45	50	2.47	66.68	0.23	17.06	2.25	76.31	2

705

706

707 Table 3. Summary of the results from the on-board measurements with $V=30 \text{ km h}^{-1}$.
708

Measurement points				Total	Total	PNC-1	GMD-1	PNC-2	GMD-2	Valid
ID	x (cm)	y (cm)	z (cm)	PNC ($\times 10^5 \#$ cm^{-3})	GMD (nm)	($\times 10^5 \#$ cm^{-3})	(nm)	($\times 10^5 \#$ cm^{-3})	(nm)	Runs
P _a	45	-45	10	73.88	73.54	6.47	21.02	67.41	82.69	4
P _b	45	0	10	2.16	42.98	0.61	13.43	1.55	67.87	2
P _c	45	45	10	0.65	59.79	0.08	11.99	0.57	100.08	4
P _d	45	-45	50	29.67	61.23	2.41	20.62	27.25	64.18	2
P _e	45	0	50	14.14	54.97	2.03	18.80	12.11	64.58	2
P _f	45	45	50	4.23	57.78	0.54	18.29	3.68	68.45	2
P _g	45	-45	90	7.91	62.24	0.81	21.03	7.10	70.60	2
P _h	45	0	90	8.07	64.92	0.73	21.22	7.34	72.68	2
P _i	45	45	90	3.37	66.65	0.41	17.62	2.96	79.93	2
P _j	80	-45	50	17.73	65.10	1.60	21.62	16.13	72.82	2
P _k	80	0	50	6.75	58.44	1.06	18.24	5.69	70.15	2
P _l	80	45	50	3.84	59.39	0.45	19.08	3.39	68.89	2

709

710 Table 4. Summary of the results from the on-board measurements with $V=40 \text{ km h}^{-1}$.
711

Measurement points				Total	Total	PNC-1	GMD-1	PNC-2	GMD-2	Valid
ID	x (cm)	y (cm)	z (cm)	PNC ($\times 10^5 \#$ cm^{-3})	GMD (nm)	($\times 10^5 \#$ cm^{-3})	(nm)	($\times 10^5 \#$ cm^{-3})	(nm)	Runs
P _a	45	-45	10	119.90	82.27	14.70	18.07	105.19	101.77	3
P _b	45	0	10	0.33	44.35	0.11	10.41	0.21	94.10	2
P _c	45	45	10	0.71	66.34	0.08	15.19	0.63	79.28	2
P _d	45	-45	50	43.16	59.25	4.19	19.45	38.96	66.70	2
P _e	45	0	50	14.88	52.60	2.32	19.56	12.56	62.95	2
P _f	45	45	50	5.79	53.45	0.89	19.12	4.90	64.18	2
P _g	45	-45	90	10.32	60.50	1.25	20.72	9.07	70.28	2
P _h	45	0	90	11.83	56.11	1.90	19.95	9.92	68.59	2
P _i	45	45	90	5.58	52.87	1.03	19.61	4.55	65.68	2
P _j	80	-45	50	26.72	55.07	4.48	19.69	22.24	68.04	2
P _k	80	0	50	8.71	50.22	2.05	19.19	6.65	63.36	2
P _l	80	45	50	3.34	57.04	4.52	18.29	2.88	68.90	2

712

Figure.1.ppt

Figure.2.ppt

Figure.3.ppt

Figure.4.ppt

Figure.5.ppt

Figure.6.ppt

Figure.8.ppt

Figure.9.ppt

Figure.10.ppt

Figure.11.ppt

Figure.12.ppt

Figure.13.ppt

Figure.14.ppt

Figure.15.ppt