1	Large eddy simulation of vehicle emissions dispersion process: Implications
2	for on-road remote sensing measurements
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#### 19 Abstract

20 On-road remote sensing technology measures concentration ratios of pollutants over CO<sub>2</sub> 21 in the exhaust plume in half a second when a vehicle passes by a measurement site, providing 22 a rapid, non-intrusive and economic tool for vehicle emissions monitoring and control. A key 23 assumption in such measurement is that the emission ratios are constant for a given plume. 24 However, there is a lack of study on this assumption, whose validity could be affected by a 25 number of factors, especially the engine operating conditions and turbulence. To guide the 26 development of the next-generation remote sensing system, this study is conducted to 27 investigate the effects of various factors on the emissions dispersion process in the vehicle 28 near-wake region and their effects on remote sensing measurement. The emissions dispersion 29 process is modelled using Large Eddy Simulation (LES). The studied factors include height of 30 the remote sensing system, vehicle speed, acceleration and side wind. The results show that the 31 measurable CO<sub>2</sub> and NO exhaust plumes are relatively short at 30 km/h cruising speed, 32 indicating that a large percentage of remote sensing readings with the measurement duration 33 of half a second are below the sensor detection limit which would distort the derived emission 34 ratio. In addition, the valid measurement region of NO/CO<sub>2</sub> emission ratio is even shorter than 35 the measurable plume and is at the tailpipe height. The effect of vehicle speed (30-90 km/h) on 36 the measurable plume length is insignificant. Under deceleration condition, the length of the 37 valid NO/CO<sub>2</sub> measurement region is shorter than those under cruising and acceleration 38 conditions. Side winds from the far-tailpipe direction have a significant effect on remote 39 sensing measurements. The implications of these findings and possible solutions to improve 40 the accuracy of remote sensing measurement have been proposed and discussed.

41 Keywords: Computational fluid dynamics; Exhaust plume dispersion; On-road emission 42 test; Near-wake region

# Abbreviations and definitions:

CFD	Computational fluid dynamics
DNS	Direct numerical simulation
LES	Large eddy simulation
LPG	Liquefied petroleum gas
PEMS	Portable emission measurement system
RANS	Reynolds-Averaged Navier-Stokes
RDE	Real driving emissions
SGS	Sub-grid scale
Emission ratio	Concentration ratio of a pollutant over CO <sub>2</sub> (e.g. NO/CO <sub>2</sub> )
Measurable exhaust plume	Concentration larger than 0.1% for CO <sub>2</sub> or 10 ppm for NO
Valid measurement region of emission ratio	Region with emission ratio close to (within $\pm 10\%$ difference) the original emission ratio in the exhaust pipe

#### 46 1. Introduction

47 On-road remote sensing technology is a rapid, non-intrusive and economic tool for use in 48 vehicle emissions monitoring and control under real driving conditions. It measures the 49 concentration ratios of pollutants (e.g. CO, HC and NO) over CO<sub>2</sub> in the exhaust plume in half 50 a second when a vehicle passes by a measurement site (Burgard et al., 2006; Cadle and 51 Stephens, 1994; Huang et al., 2018c). These emission ratios are highly useful parameters to 52 evaluate the emissions performance of a vehicle and have been used in various applications 53 worldwide. Since 1 September 2014, the Hong Kong Environmental Protection Department 54 (HKEPD) started using on-road remote sensing technology to detect high-emitting petrol and 55 liquefied petroleum gas (LPG) vehicles for enforcement (HKEPD, 2018). The enforcement 56 program has been very effective in reducing the emissions from petrol and LPG vehicles 57 (Huang et al., 2018b; Organ et al., 2019). However, diesel vehicles, a major source of NO<sub>x</sub> 58 emissions, are excluded because the current remote sensing system would likely produce 59 significant false detections of diesel high-emitters. The underlying reasons include low 60 pollutant concentrations and large CO<sub>2</sub> variations in the exhaust plume of diesel vehicles due 61 to their non-premixed lean combustion mode. To extend the enforcement program to diesel 62 vehicles, a new generation of remote sensing system with higher accuracy is under development (Huang et al., 2018b). 63

A key assumption in remote sensing measurement is that the emission ratios are constant for a given plume (Bishop *et al.*, 1989; Burgard *et al.*, 2006). However, there is a lack of study on this assumption, whose validity could be affected by a number of factors such as the engine conditions and vehicle-, exhaust- and wind-induced turbulence. In addition, remote sensing system is placed at a fixed height above the road to measure the emissions of all the passing vehicles, e.g. typically 25 cm for vehicles with bottom arranged tailpipes. However, the tailpipe height varies among vehicle models, as well as vehicle loads. The alignment of the remote sensing light beam to the vehicle tailpipe determines the amount of exhaust gas being measured
and thus affects the remote sensing accuracy.

73 Measuring emissions in the near-wake region of a fast-moving vehicle is challenging. This 74 is because the instantaneous tailpipe emissions in a real driving task are highly dynamic. 75 Remote sensing scans the exhaust plume from tailpipe exit to a few meters downstream in half 76 a second at a frequency of 100 or 200 Hz. In comparison, other emission measurement 77 techniques cannot achieve the required time resolution or measurement region under real 78 driving conditions, and thus are not suitable for investigation of emissions dispersion process 79 for remote sensing applications. Portable emissions measurement system (PEMS) is a standard 80 method to measure real driving emissions (RDE) by carrying measurement instruments on-81 board the target vehicle (Degraeuwe and Weiss, 2017; Vlachos et al., 2014). However, the time 82 resolution of PEMS is 1 Hz and can only measure the emissions in the tailpipe before dispersion. 83 Plume chasing is a technology that measures the emissions in the wake of a target vehicle by 84 using a following laboratory vehicle carrying measurement instruments (Lau et al., 2015; Ning 85 et al., 2012). However, it can only measure the emissions in the far downstream region (10-15 86 m) due to safety issues. In addition, the time resolution is only 1-10 seconds and the 87 measurement height (1.5 m) is above the tailpipe. On the other hand, computational fluid 88 dynamics (CFD) modelling is a feasible and economic tool to provide detailed and visualised 89 information about the turbulent flows. It can help the design and development of more accurate 90 remote sensing devices, e.g. for diesel high-emitters enforcement program.

To numerically simulate the turbulence fields, a set of partial differential equations for continuity, momentum (i.e. Navier-Stokes equations), energy and thermodynamic variables need to be solved (Elger *et al.*, 2012). Unfortunately, analytical solutions for the Navier-Stokes equations are not available, except for a few very simple conditions. Therefore, numerical methods are usually used to solve the governing equations. Currently, the most popular

96 numerical approach is the Reynolds-Averaged Navier-Stokes (RANS). In RANS, the local 97 fluctuations and turbulence structures are integrated in the mean quantities. To close the 98 Reynolds stress term, the most widely used models are the k- $\varepsilon$  and k- $\omega$  two-equation models 99 (Argyropoulos and Markatos, 2015). Chan et al. (2001) simulated the initial dispersion process 100 of NO<sub>x</sub> from a vehicle exhaust plume using a k- $\varepsilon$  turbulence model, and observed that the effect 101 of exhaust-induced turbulence was more dominant than wind-induced turbulence in the vicinity 102 of tailpipe exit. In comparison, wind-induced turbulence gradually became significant for  $NO_x$ 103 dispersion with the development of exhaust plume. Amorim et al. (2013) evaluated the impact 104 of urban trees on the dispersion of road traffic CO using a standard k- $\varepsilon$  turbulence model. The 105 results showed that local air quality was strongly dependent on the synergies between the 106 meteorological conditions, street canyon configuration and trees. Bhautmage and Gokhale 107 (2016) investigated the effects of moving-vehicle wakes on pollutant dispersion in a highway 108 road tunnel by realizable k- $\varepsilon$  model. Woo *et al.* (2016) investigated the influence of a leading 109 vehicle's exhaust plume on a following mobile laboratory vehicle's air quality measurements 110 using both PEMS experiments and CFD simulation (standard k- $\varepsilon$  model).

111 However, the modelling ability of RANS is only limited to the large flow structures. With 112 the fast increase of computational power, more advanced turbulence models are becoming 113 feasible in recent years, such as Large Eddy Simulation (LES) and Direct Numerical 114 Simulation (DNS). DNS solves all the turbulence fluctuations and structures without averaging 115 or using turbulence models, providing the most accurate numerical solution to turbulence flows. 116 However, DNS is only applicable to fundamental research applications with low Reynolds 117 numbers, and thus is not applicable to most practical applications due to its tremendous 118 requirement for computer speed and memory (Argyropoulos and Markatos, 2015; Elger et al., 119 2012). LES is a compromise between RANS and DNS (Spalart, 2000), in which the larger-120 scale flow structures are explicitly computed while the effect of sub-grid scale (SGS) motions

121 is modelled (Veynante and Vervisch, 2002). LES offers the ability to see more detailed flow 122 structures (Spalart, 2000). In view of the limited computational resource, LES is more 123 applicable for flow fields with a moderate Reynolds number and geometry size (Rutland, 2011). 124 Several studies have utilised LES to simulate the emissions dispersion process. Dong and Chan 125 (2006) and Chan et al. (2008) used LES to simulate the dispersion of NO<sub>x</sub> and CO in the near-126 wake region of a light-duty diesel vehicle. Gallagher et al. used LES to investigate the effect 127 of various factors on the dispersion of gaseous emissions from the street to footpath region in 128 a street canyon, including passive control system (e.g. parked cars, low boundary walls) 129 (Gallagher et al., 2011, 2013), wind conditions and fleet composition (Gallagher, 2016), and 130 geometrical details of parked cars and mesh size (Gallagher and Lago, 2019). Aristodemou et 131 al. (2018) used an LES model to investigate the effect of tall buildings on air flows and 132 pollutants dispersion.

133 As reviewed, on-road remote sensing is considered an effective and economic tool to 134 monitor emissions of vehicle fleets in the real world. However, there is a lack of investigation 135 on the fundamental assumption of its measurement principle, and a thorough understanding of 136 the tailpipe emissions dispersion process is of great importance for the development of the 137 next-generation remote sensing system for diesel high-emitters enforcement. Therefore, this 138 study is conducted to unprecedentedly investigate the effect of various factors on emissions 139 dispersion process in the vehicle near-wake region using LES, with a special focus on its effect 140 on emissions measurement using on-road remote sensing technology. The implications of the 141 CFD results and possible solutions to improve the accuracy of remote sensing measurement 142 are discussed.

#### 144 **2.** Numerical models

### 145 *2.1. Computational mesh*

146 Fig. S1 (Supplementary Material) shows the computational domain (a), dimensions of the 147 vehicle body (b) and computational mesh (c). As shown in Fig. S1(a), the computational domain is a rectangular box of  $20 \text{ m} \times 6 \text{ m} \times 5 \text{ m}$ . The studied vehicle is a diesel-fuelled Toyota 148 149 HiAce with an engine displacement of 2982 cc (Fig. S1(b)), which is placed at 4 m in the 150 computational domain. Toyota HiAce is the most popular diesel vehicle model in Hong Kong, 151 accounting for 19% of total remote sensing records (Huang et al., 2018a). The computational 152 mesh is generated using the ANSYS Meshing. As shown in Fig. S1(c), the computational mesh 153 mainly consists of tetrahedral grids. It is generated according to the best practice guideline for 154 flows in the urban environment (Franke et al., 2007; Gallagher et al., 2011). The general grid 155 size is 0.10 m. However, the grids in the near-wake region of the vehicle (the area of the main 156 interest) are refined to 0.05 m. The grids around the vehicle surface, exhaust pipe and ground 157 are also refined to capture the flow characteristics around the boundary layer and exhaust pipe. 158 The total number of nodes is 1384083.

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#### 160 *2.2. Computational model*

161 The numerical model is developed using the CFD code ANSYS Fluent 19.2 (ANSYS, 2019). In this study, the Reynolds numbers ( $Re = \rho uL/\mu$ ) are in the ranges of  $2.5 \times 10^5$  – 162  $3.2 \times 10^6$  and  $1.5 \times 10^4 - 4.8 \times 10^4$  for air and exhaust gas flows, respectively. The ambient air and 163 exhaust gas flows are simulated using the LES approach. The SGS flows are simulated using 164 165 the Wall-Adapting Local Eddy-Viscosity (WALE) model which provided several advantages 166 over the classical Smagorinsky formulation, including the detection of all turbulence structures 167 relevant for kinetic energy dissipation, correct wall asymptotic behaviour for wall bounded 168 flows, and correct treatment of laminar zones in the domain (Nicoud and Ducros, 1999). Mass,

169 energy and momentum conservation, heat transfer, and species transfer equations are 170 numerically solved. Diesel vehicles are relatively clean in CO and HC emissions (Huang et al., 171 2019a; Huang et al., 2019b) but are the major sources of NO pollution in cities (Anenberg et 172 al., 2017). Therefore, five gas species including N<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>O, CO<sub>2</sub> and NO are considered. NO 173 is considered an inert species in this study because remote sensing only measures the exhaust 174 emissions from the tailpipe exit to a few meters downstream in half a second which is relatively short compared with the time scales of NO<sub>x</sub>-O<sub>3</sub> reactions (tens of seconds) (Vardoulakis et al., 175 176 2003). The pressure-velocity coupling scheme is the Semi-Implicit Method for Pressure Linked 177 Equations (SIMPLE) method. The spatial discretization methods are Least Squares Cell Based 178 for gradient, Second Order for pressure, Bounded Central Differencing for momentum and 179 Second Order Upwind for energy and species transport. Transient simulations are carried out with a time step increment of 0.01 s. The convergence criteria are residual values of  $10^{-3}$  for 180 continuity, velocity and species, and 10<sup>-6</sup> for energy. The maximum number of iterations is 30 181 182 per time step. 1000 steps are performed to obtain reliable and developed exhaust plumes. Fig. 183 S2 shows the temporal variation of  $CO_2$  concentrations at different distances downstream the 184 tailpipe exit. It shows that the CO<sub>2</sub> concentration becomes relatively stable after 200 steps, 185 although with slight variations. Therefore, the instantaneous images shown are representative 186 of the pollutant dispersion in the near-wake region of a vehicle. In Section 3, the plume lengths 187 are the averaged values between time steps 250 and 1000.

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### 2.3. Boundary and initial conditions

In this study, various vehicle speed, acceleration and wind conditions are simulated because they are the primary parameters determining the exhaust plume conditions and the flow fields in the vehicle near-wake region, which affect the emissions dispersion process and consequently the remote sensing measurement. **Table S1** (Supplementary Material) presents 194 the boundary and initial conditions used in the simulation. In all the simulated cases, the 195 ambient air temperature is 300 K and standard air compositions are used. Vehicle speed and 196 acceleration are the key parameters that determine the engine operation conditions and thus the 197 tailpipe emissions, which are also the key parameters that affect the flow fields around the 198 vehicle (vehicle and exhaust induced turbulence). In this study, four vehicle speeds (7, 30, 60 199 and 90 km/h) and three accelerations (-3, 0, +3 km/h/s) are simulated. 7 km/h is the lowest 200 speed that can be measured in remote sensing and 90 km/h is the highest speed tested in the 201 Hong Kong Transient Emission Test (HKTET), which is the test cycle for ascertaining the 202 rectification of the excessive emission problem of in-use vehicles with the aid of a chassis 203 dynamometer. Therefore, 7-90 km/h is the range of driving conditions that is used in remote 204 sensing analysis. Side wind is another key parameter that affects the flow field around the 205 vehicle (wind induced turbulence). In this study, seven side wind speed conditions are 206 simulated, namely  $0, \pm 2, \pm 6$  and  $\pm 12$  m/s which correspond to calm (Beaufort scale 0), light 207 breeze (Beaufort scale 2), moderate breeze (Beaufort scale 4) and strong breeze (Beaufort scale 208 6), respectively.

209 To obtain boundary conditions at the tailpipe exit, a RDE test on a Toyota HiAce was 210 conducted using PEMS. During the RDE test, the vehicle speed, exhaust flow rate, exhaust gas 211 concentrations and exhaust temperature were measured at a frequency of 1 Hz. More details 212 about the PEMS equipment can be found in Huang et al. (2019c). Fig. S3 shows the driving 213 speed, CO<sub>2</sub> concentration and exhaust temperature profiles of the RDE test. As shown in Fig. 214 S3, a RDE test is highly dynamic. Therefore, the average values of exhaust flow rate, 215 temperature and concentrations were calculated from the RDE data under each simulated 216 vehicle speed and acceleration condition. The exhaust gas speed at the tailpipe exit is calculated 217 from the tailpipe diameter of 63.5 mm (2.5 inches) and the PEMS data of exhaust flow rate and temperature. A high-emitting event of NO/CO<sub>2</sub> = 50 ppm/% is simulated for all cases. 218

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# 2.4. Mesh independence and model verification

221 Mesh quality is an important concern in CFD modelling. To achieve mesh independence, 222 three meshes with different grid sizes have been tested, namely Mesh 1 (816900 nodes), Mesh 2 (1384083 nodes) and Mesh 3 (1840211 nodes). Fig. 1 compares the averaged CO<sub>2</sub> 223 224 concentration along the tailpipe centreline as a result of different mesh densities. Compared 225 with Mesh 2 (medium mesh), Mesh 1 (coarse mesh) over-predicts  $CO_2$  concentration within 226 0.15 m but under-predicts it in the further downstream. When the mesh is refined from Mesh 227 2 to Mesh 3 (fine mesh), the two profiles become almost identical within 0.2 m, but Mesh 3 228 slightly over-predicts it between 0.2 and 3.0 m. In general, the difference between the two 229 calculated profiles becomes relatively small, indicating that mesh independence is achieved. 230 The computation time using Mesh 2 is about 40 hours per run using 24 cores of a 2.3 GHz Intel 231 Xeon E5-2695 v3 workstation, while the computation time of Mesh 3 is 70% longer than that 232 of Mesh 2. Therefore, Mesh 2 is considered sufficient to perform the simulations with a 233 reasonable accuracy and low computational cost.

234 To verify the CFD model, a diesel Toyota HiAce vehicle was tested under idling condition 235 to obtain the CO<sub>2</sub> concentration and gas temperature distributions downstream the tailpipe. CO<sub>2</sub> 236 concentration was measured by an EMS 5003 gas analyser and gas temperature was measured 237 by a K-type thermocouple. As shown in Fig. 2, the model well predicts CO<sub>2</sub> concentrations 238 and exhaust temperature within 0.3 m and after 3.0 m, but over-predicts them between 0.3 and 239 3.0 m. Although CO<sub>2</sub> is over-predicted at some region, it will not under-estimate the NO/CO<sub>2</sub> 240 ratio because the emissions dispersion process is in essence the mixing of exhaust plume, which 241 carries the emissions, with the ambient air, so that CO<sub>2</sub>, NO and temperature show the same 242 distribution pattern in the region close to the tailpipe exit (Fig. S4). This indicates that NO will 243 also be over-predicted to the same extent as CO<sub>2</sub> and temperature (Fig. 2), thus NO/CO<sub>2</sub> ratio

244 will not be under-predicted. This also justifies the validation method using temperature and 245 CO<sub>2</sub> which can be easily and reliably measured. The over-prediction of CO<sub>2</sub> and temperature 246 would be caused by the simplification of the vehicle geometry, in which the tailpipe is 247 horizontal while it is slightly bent towards the ground in a real HiAce. The over-prediction in the medium distance downstream was also reported by Chan et al. (2001). In the real world, 248 249 some rear-arranged tailpipes are with a slight bent exit, which would lead to shorter measurable 250 exhaust plumes at the tailpipe height than horizontal tailpipes, if the other conditions (mainly 251 exhaust flow rate and gas concentrations) are the same. A shorter plume will be more 252 challenging for remote sensing measurement as remote sensing sensors are placed at a fixed 253 height above road surface, which will be further discussed in Fig. 4. Overall, the fast dispersion 254 of the experimental exhaust plume, as a key phenomenon, has been well captured by the LES 255 model.

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## 257 **3.** Results and discussion

258 The CFD results are presented and discussed as follows. Section 3.1 reports exhaust plume 259 dispersion process at vehicle speed of 30 km/h, acceleration of 0 km/h/s and side wind of 0 m/s. 260 The distributions of air velocity, dynamic pressure, temperature, emission concentrations and 261 emission ratios are visualised, and the effects of emissions dispersion and tailpipe height on 262 remote sensing measurement are analysed. Sections 3.2, 3.3 and 3.4 investigate the effects of 263 vehicle speed, acceleration and side wind on emissions dispersion process and remote sensing 264 measurement, respectively. 30 km/h is used as the base vehicle speed because it is the typical 265 speed when vehicles pass by a remote sensing site (Fig. S5).

#### 3.1. Emissions dispersion process

Fig. 3 shows the velocity (a), dynamic pressure (b) and temperature (c) fields around the vehicle on the vertical and horizontal cut-planes that pass through the tailpipe centreline. The results demonstrate that the air flow speed is slow in the vehicle near-wake region (Fig. 3(a)), leading to a low dynamic pressure ( $q = \frac{1}{2} \rho u^2$ ) region (Fig. 3(b)). The exhaust temperature reduces quickly to ambient temperature in a very short distance after the tailpipe exit (Fig. 3(c)). In general, the effect of the exhaust gas flow on the overall air flow fields of velocity, dynamic pressure and temperature is insignificant due to its small mass.

275 Fig. 4 visualises the distribution of the absolute volume concentrations of  $CO_2$  (a) and NO 276 (b) emissions in the vehicle near-wake region. Areas with concentrations lower than 0.1% for 277 CO<sub>2</sub> and 10 ppm for NO are clipped to demonstrate the exhaust plume length, which are about the typical detection limits of CO<sub>2</sub> and NO in PEMS, respectively (Horiba, 2017), while the 278 279 detection limits of remote sensing system are much higher than PEMS (Huang et al., 2018b). 280 Therefore, the measurable exhaust plume is defined as the main plume (excluding the detached 281 discrete plume areas) with emission concentration larger than 0.1% for CO<sub>2</sub> or 10 ppm for NO. 282 Using this definition, Fig. 4 shows that the CO<sub>2</sub> plume lengths are  $2.7 \pm 1.1$  m in the top vertical 283 plane and  $1.5 \pm 0.2$  m in the bottom horizontal plane, and the NO plume lengths are  $1.4 \pm 0.2$ 284 and  $1.1 \pm 0.3$  m, respectively. In comparison, NO plume is smaller than CO<sub>2</sub> plume due to its 285 much lower concentration (at ppm level) than that of CO<sub>2</sub> (at % level) in the engine exhaust 286 gas.

However, a remote sensing measurement lasts for half a second at 200 Hz when a vehicle passes by the measurement site, which is equivalent to 4.2 m at the studied vehicle speed of 30 km/h. **Fig. 4** indicates that at least 36%-74% of the remote sensing measurements are meaningless for a given plume, because the CO<sub>2</sub> and NO concentrations are below the sensitivity of a remote sensor after 1.1-2.7 m. For each exhaust plume, remote sensing records 292 100 readings of NO and CO<sub>2</sub> emission concentrations, and plots NO against CO<sub>2</sub> to derive the 293 least square slope, i.e. NO/CO<sub>2</sub> ratio. The large percentage of meaningless data points (around 294 the origin, as shown in Fig. 2 of Bishop *et al.* (1989)) would distort the derived emission ratio, 295 and thus reduce the accuracy of remote sensing. This suggests that using a shorter measuring 296 duration and a higher remote sensing frequency could improve the accuracy.

297 As an indirect measurement technique, remote sensing can only determine the 298 concentration ratio of NO/CO<sub>2</sub> rather than the absolute concentration of NO or CO<sub>2</sub>, and a key 299 assumption is that NO/CO<sub>2</sub> ratio is constant for a given plume. Fig. 5 shows the distribution of 300 NO/CO<sub>2</sub> ratio in the vertical (top) and horizontal (bottom) cut-planes passing through the 301 tailpipe centreline. In this study, the NO/CO<sub>2</sub> value is 50 ppm/% at the tailpipe exit. However, 302 as shown in Fig. 5, the assumption that NO/CO<sub>2</sub> ratio is constant is only valid within a very 303 short distance due to the different dispersion rates of NO and CO<sub>2</sub> in the further downstream 304 region. This is because the exhaust gas speed is high in the region close to the tailpipe exit, so 305 that the mixing and dispersion are mainly controlled by the high momentum of the exhaust gas. 306 As a result, NO and CO<sub>2</sub> disperse at the same rate, leading to the unchanged NO/CO<sub>2</sub> ratio near 307 the tailpipe exit. This is proven by the results shown in Fig. S4 that CO<sub>2</sub> and NO emissions 308 demonstrate the same distribution as the exhaust temperature in the region near the tailpipe exit 309 (within  $\sim 1$  m). However, in the further downstream region, the exhaust gas speed and 310 momentum reduce quickly, so that the molecular diffusion process becomes the dominant 311 factor. At ambient temperature of 300 K, the diffusion coefficients in air are 15.84 mm<sup>2</sup>/s for CO<sub>2</sub> and 23.69 mm<sup>2</sup>/s for NO, respectively (Yaws, 2003, 2010). Therefore, NO disperses more 312 313 quickly than CO<sub>2</sub> does, leading to the reduced NO/CO<sub>2</sub> ratio in the further downstream region. 314 NO/CO<sub>2</sub> ratio is 50 ppm/% at the tailpipe exit but then reduces gradually with the 315 development of the exhaust plume. To quantify the analysis, the valid measurement region of 316 emission ratio is defined as regions with emission ratio close to (within  $\pm 10\%$  difference) that of the original emission ratio in the exhaust pipe, i.e. 45-50 ppm/% in this study. Using this definition, the lengths of the valid measurement region for remote sensing are only about 1.2  $\pm 0.4$  m in the vertical plane and  $1.1 \pm 0.3$  m in the horizontal plane. The valid measurement region of emission ratio is even shorter than that of measurable exhaust plume. This further indicates that the currently used remote sensing measurement duration of half a second for one passing vehicle is too long and would cause additional uncertainties.

323 In addition, remote sensing systems are placed at a fixed height to measure emissions of 324 all the passing vehicles. However, tailpipe height varies among vehicle models, as well as 325 vehicle loads. Fig. 5 shows that the valid measurement region of NO/CO<sub>2</sub> ratio is only at the 326 tailpipe height. Remote sensing system would measure no emissions if the sensor is lower than 327 the tailpipe, and would measure much lower NO/CO<sub>2</sub> ratios if the sensor is higher than the 328 tailpipe. To more accurately measure the emissions from different vehicles, one possible 329 solution is using multiple remote sensors with different vertical heights in one remote sensing 330 system, and only use data from the sensor that has the strongest signals, which is more likely 331 aligned with the tailpipe centreline. Another promising method is a facing-down remote 332 sensing configuration which is currently under development.

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*334 3.2. Effect of vehicle speed* 

Fig. 6(a) shows the effect of vehicle speed on the measurable CO<sub>2</sub> plume length (> 0.1% concentration) on the vertical cut-plane crossing the tailpipe centreline, including 7, 30, 60 and 90 km/h. Table S2 provides the lengths of the measurable CO<sub>2</sub> and NO/CO<sub>2</sub> plumes as a function of the vehicle speed. As shown in Fig. 6(a), at 7 km/h vehicle speed, the turbulence included by the vehicle is weak and does not significantly affect the exhaust plume. As a result, the exhaust plume is slim and long, with a measurable CO<sub>2</sub> plume length of  $3.9 \pm 1.9$  m. The plume is generally distributed horizontally at the tailpipe height. Higher vehicle speed induces 342 stronger turbulence and thus enhances the mixing and dispersion processes of exhaust 343 emissions. In addition, higher vehicle speed induces some vortices in the near wake region, 344 which entrain some exhaust gas into the near-wake region above the tailpipe. Consequently, 345 the measurable  $CO_2$  plume lengths of 30-90 km/h vehicle speeds are significantly shorter than 346 that of 7 km/h, but they remain relatively constant from 30 to 90 km/h (with  $2.7 \pm 1.1 \text{ m}$  @ 30 347 km/h,  $2.1 \pm 0.4 \text{ m}$  @ 60 km/h and  $2.1 \pm 0.2 \text{ m}$  @ 90 km/h).

348 Regarding the length of the valid measurement region of NO/CO<sub>2</sub> ratio (i.e. 45-50 ppm/%), 349 Fig. 6(b) shows that it generally reduces with increasing vehicle speed, with  $1.9 \pm 0.4$  m at 7 350 km/h,  $1.2 \pm 0.4$  m at 30 km/h,  $0.7 \pm 0.1$  m at 60 km/h and  $1.0 \pm 0.4$  m at 90 km/h. It is worth 351 noting that the measurable plume lengths (2.1-3.9 m, Fig. 6(a)) are always longer than the valid 352 measurement region of NO/CO<sub>2</sub> (0.7-1.9 m, Fig. 6(b)). However, both lengths are critical for 353 remote sensing: the measurable plume length determines the region in which meaningful 354 remote sensing readings can be made, while the valid measurement length determines the 355 region in which remote sensing readings can be used to derive the correct NO/CO<sub>2</sub> ratio. The 356 shorter one limits the plume length that can be used for remote sensing measurement.

357 In a real-world remote sensing program, 7 km/h is the lower measurement limit of vehicle 358 speed and most vehicles are driven faster than 7 km/h. Higher vehicle speed induces stronger 359 turbulence which enhances exhaust plume dispersion. On the other hand, higher vehicle speed requires bigger engine load and thus produces higher exhaust flow rate which prolongs the 360 361 exhaust dispersion. Therefore, the effect of vehicle speed (30-90 km/h) on the lengths of 362 measurable CO<sub>2</sub> plume and valid NO/CO<sub>2</sub> measurement region is not obvious. This finding 363 suggests that vehicle speed would not significantly affect the remote sensing measurement. 364 High vehicle speed like 90 km/h would even be better for a reliable remote sensing measurement as the engine produces sufficient exhaust gas. 365

368 Fig. 7(a) shows the effect of vehicle acceleration on the measurable CO<sub>2</sub> plume length (> 369 0.1% concentration), including moderate deceleration of -3 km/h/s, cruising (0 km/h/s) and 370 moderate acceleration of +3 km/h/s. PEMS data show that under the same vehicle speed 371 condition (i.e. 30 km/h), there are insignificant differences in the exhaust temperature and CO<sub>2</sub> concentration between deceleration (534 K and 6.1%), cruising (548 K and 9.0%) and 372 373 acceleration (567 K and 9.1%) for the studied HiAce diesel vehicle. The major differences are in the exhaust flow rates, with 32.8, 49.3 and 135.0 L/s for deceleration, cruising and 374 375 acceleration, respectively, due to higher fuel consumption and engine speed at higher load. 376 Therefore, as shown in Fig. 7(a), the measurable  $CO_2$  plume length under deceleration 377 condition  $(1.6 \pm 0.5 \text{ m})$  is shorter than that under cursing  $(2.7 \pm 1.1 \text{ m})$  or acceleration  $(4.6 \pm 1.1 \text{ m})$ 1.9 m) condition. Such a short plume would be too challenging to be measured by remote 378 379 sensing and the derived least square slope (i.e. NO/CO<sub>2</sub>) would include high uncertainties. 380 Regarding the valid measurement region of emission ratio for remote sensing, Fig. 7(b) shows 381 that the valid measurement region with NO/CO<sub>2</sub> ratio between 45 and 50 is relatively short 382 under deceleration condition  $(0.7 \pm 0.1 \text{ m})$  compared with acceleration  $(2.3 \pm 0.5 \text{ m})$ . These 383 results suggest that remote sensing measurements under deceleration conditions would have 384 larger uncertainties and should be used with caution. Fuel injection would be even stopped 385 (thus no exhaust emissions) under strong deceleration conditions, e.g. typically < -5 kW/t vehicle specific power (VSP) for passenger cars (Bernard et al., 2018). Therefore, it is 386 387 recommended to use remote sensing measurements under steady speed or moderate 388 acceleration conditions which generate sufficient amount of exhaust plumes for accurate 389 measurements.

392 Fig. 8 shows the effect of side wind on the CO<sub>2</sub> plume and NO/CO<sub>2</sub> ratio on a horizontal 393 cut-plane passing through the tailpipe centreline, including calm (0 m/s), light breeze ( $\pm 2$  m/s, 394 Beaufort scale 2), moderate breeze ( $\pm 6$  m/s, Beaufort scale 4) and strong breeze ( $\pm 12$  m/s, Beaufort scale 6). Side winds from different directions are simulated, where "+" indicates that 395 396 the side wind comes from the tailpipe side of the vehicle while "-" indicates that the side wind 397 comes from opposite side. The arrows in Fig. 8 indicate the relative air velocity vectors to the 398 vehicle. As shown in Fig. 8, when side wind speed increases from 0 to +12 m/s, CO<sub>2</sub> plume 399 bends with the side wind and the measurable CO<sub>2</sub> plume length does not change significantly. 400 However, when side wind speed increases from 0 to -12 m/s, the measurable CO<sub>2</sub> plume 401 becomes very short even under light breeze condition of -2 m/s. This is mainly because when 402 the side wind comes from the far-tailpipe side, the relative air velocity vector has an angle with 403 the vehicle body. As a result, the vehicle body blocks the wind and generates vortices (i.e. 404 stronger turbulence) in the near-wake and leeward region. These vortices entrain the exhaust 405 plume to the upper right region in the vehicle near wake region, leading to the greatly reduced 406 plume lengths observed in Fig. 8. Regarding the valid measurement region of NO/CO<sub>2</sub> ratio 407 (45-50 ppm/%), Fig. 8 shows that it reduces significantly with the increase of far-tailpipe side 408 winds, but not obviously for tailpipe side winds. These results suggest that side winds from the 409 far-tailpipe direction would have a significant effect on remote sensing measurement in terms 410 of measurable CO<sub>2</sub> plume and valid NO/CO<sub>2</sub> region, and thus remote sensing measurements 411 should ideally be taken under calm conditions.

412

#### 413 **4.** Conclusions

414 In this study, LES modelling was performed to assess the effects of various factors on 415 emissions dispersion process in the vehicle near-wake region and their effects on emissions 416 measurement using on-road remote sensing technology. The simulated factors included remote 417 sensing height, vehicle speed, acceleration and side wind. The major results are summarised as 418 follows:

419 The lengths of measurable CO<sub>2</sub> and NO plumes and valid NO/CO<sub>2</sub> measurement region 1) 420 are relatively short at 30 km/h cruising speed, suggesting that the currently used 421 measurement duration of half a second would be too long. Using a higher remote sensing 422 frequency and a shorter measurement duration should improve the accuracy. In addition, 423 the valid NO/CO<sub>2</sub> measurement region is at the tailpipe height, so that measurement would 424 be inaccurate if the remote sensing beam is not well aligned with the tailpipe. A downward-425 facing configuration or an array of beams with different heights could help improve remote 426 sensing accuracy.

427 The exhaust plume is slim and long at 7 km/h vehicle speed, but becomes significantly 2) 428 shorter at higher vehicle speeds. The effect of vehicle speed (30-90 km/h) on the lengths 429 of measurable CO<sub>2</sub> plume and valid NO/CO<sub>2</sub> measurement region is insignificant, due to 430 the competing effects of stronger vehicle induced turbulence and higher exhaust flow rate. 431 3) Under deceleration condition (-3 km/h/s), the measurable CO<sub>2</sub> plume and the valid 432 NO/CO<sub>2</sub> measurement region are shorter than those under cruising (0 km/h/s) and 433 acceleration (+3 km/h/s) conditions due to lower exhaust flow rate. This suggests that 434 remote sensing measurements should be taken under steady speed or moderate acceleration 435 conditions which generate sufficient amount of exhaust plumes for accurate measurements. 436 4) Far-tailpipe side winds reduce the lengths of the measurable plume and valid NO/CO<sub>2</sub> 437 measurement region, especially for those from the far-tailpipe side. Therefore, remote

sensing should ideally be used under calm conditions.

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Fig. 1. Mesh independence test.





**Fig. 2**. Comparison of simulated and measured CO<sub>2</sub> concentrations and gas temperatures.



**Fig. 3.** Velocity (a), dynamic pressure (b) and temperature (c) around the vehicle under

vehicle speed of 30 km/h, acceleration of 0 km/h/s and side wind of 0 m/s.

553



555 Fig. 4. Absolute volume concentrations of CO<sub>2</sub> (a) and NO (b) under vehicle speed of 30





- **Fig. 5.** Concentration ratio of NO/CO<sub>2</sub> under vehicle speed of 30 km/h, acceleration of 0
- 559 km/h/s and side wind of 0 m/s.



561 Fig. 6. Absolute volume concentrations of  $CO_2$  (a) and concentration ratios of NO/CO<sub>2</sub> (b)

under different vehicle speed conditions. Acceleration is 0 km/h/s and side wind is 0 m/s.



564 Fig. 7. Absolute volume concentrations of  $CO_2$  (a) and concentration ratios of  $NO/CO_2$  (b)

under different acceleration conditions. Vehicle speed is 30 km/h and side wind is 0 m/s.



567 Fig. 8. Absolute volume concentrations of CO<sub>2</sub> and concentration ratios of NO/CO<sub>2</sub> under
568 different side wind conditions. Vehicle speed is 30 km/h and accelerations is 0 km/h/s.