Effect of Sampling Duration on the Estimate of Pollutant Concentration Behind a Heavy-Duty Vehicle: a Large-Eddy Simulation

Jingwei Xie¹, Chun-Ho Liu¹, Yuhan Huang^{2,3}, Wai-Chuen Mok^{1,2,3}

¹Department of Mechanical Engineering, The University of Hong Kong, Hong Kong

²School of Civil and Environmental Engineering, University of Technology Sydney, Australia

³Jockey Club Heavy Vehicle Emission Testing & Research Centre, Vocational Training Council, Hong Kong

Highlights

- Large-eddy simulation is conducted for the tailpipe dispersion of an on-road truck
- Fluctuating concentrations are tightly related to the turbulence in the near wake
- Sampling accuracy is affected by sampling duration and fluctuating concentrations
- Concentration reading is more accurate if sampling points are closer to the tailpipe
- Sampling at dominant frequencies is necessary to reduce sampling uncertainty

 sampling accuracy. With a longer sampling (averaging) duration, the sample mean concentration converges to the population mean, improving the sample reliability. However, this effect is less pronounced in long sampling duration. The sampling accuracy is also 30 influenced by the locations of sampling points. For the region $x > 0.6h$, the sampling accuracy is degraded to a large extent. As a result, acceptable sample mean is hardly achievable. Finally, frequency analysis unveils the mechanism leading to the variance in concentration measurements which is attributed to sampling duration. Those data with frequency higher than the sampling frequency are filtered out by moving average in the statistical analyses.

 (283 words) *Keywords:* Measurement uncertainty; plume chasing; plume meandering; sampling reliability; tailpipe dispersion; turbulent wake

1. Introduction

 Vehicular exhaust consists of greenhouse gases and toxic pollutants that would lead to various detrimental health concerns, including respiratory symptoms, disease and cancer (Smit et al., 2019; Tayarani & Rowangould, 2020). In Hong Kong, road transport contributed 20%, 43 53%, 10% and 13% to the annual emissions of nitrogen oxides (NO_X) , carbon monoxide (CO) , 44 respirable suspended particulate (PM_{10}) and fine suspended particulate $(PM_{2.5})$, respectively (HKEPD, 2019). Vehicular pollutant is crucial to pedestrian-level air quality because of its close proximity to stakeholders (Smit et al., 2019). Hence, roadside pollutant concentrations are usually much higher compared with ambient ones. The impact is more severe in cities due to huge population. Most air-pollution-related premature deaths are pertinent to vehicular exhaust (Caiazzo et al., 2013). It was estimated that vehicular exhaust resulted in 385,000 premature deaths and around US\$ 1 trillion in health damages worldwide in 2015 (Anenberg et al. 2019). In recent years, the increasing traffic volumes and high-rise, dense buildings further worsen the roadside air pollution problem (Huang et al., 2021). Thus, proper control of vehicular exhaust, in particular the reliable identification of heavy on-road emitters, should be enacted.

 In view of depreciation, inappropriate maintenance, tampering or breakdown of engine components, in-use vehicles often violate emission regulations (Huang et al., 2020a). Road conditions, such as slope and traffic congestion, influence the emission directly (Davison et al., 2020; Smit & Kingston, 2019), which, however, are hardly modelled in laboratories. In this connection, on-road sampling techniques, including portable emission measurement system (PEMS), on-road remote sensing, exhaust plume chasing, together with tunnel and roadside ambient measurements, have been developed (Huang et al. 2018). Tunnel and roadside ambient measurements are designed for group sampling but not individuals (Smit et al., 2010). Remote sensing is a non-intrusive way to identify heavy on-road emitters. However, its functionality is degraded by the short sampling episode (less than the turbulence timescale, seconds, in vehicular wakes) and the constraints of sampling locations (Wu et al. 2017). Among others, PEMS and plume chasing enable long-term (minutes) emission-data collection for a specific vehicle. The applicability of PEMS for fleetwide measurements is limited by its long turnover time (Franco et al., 2013). Practically, plume chasing realizes the on-road measurements of individual vehicles (Ježek et al., 2015). Another vehicle, which is equipped with rapid-response pollutant analysers, followsthe target vehicle for (continuous) data collection during real-world driving conditions. Plume chasing is high throughput (compared with PEMS), facilitating massive on-road data collection for vehicle-fleet exhaust and emission technology (Wang et al. 2020). In view of road safety, a minimum separation is required between the two vehicles (roughly 10 m). Nonetheless, this shortcoming can be overcome by towing a mobile laboratory after the targeted vehicle (Morawska et al., 2007).

 Implementation of plume chasing, on the other hand, is complicated by the turbulent wake behind the target vehicle (Yang et al. 2018). After tailpipe exhaust, the plume undergoes dilution in two regimes (Morawska et al. 2007). Within the near-wake regime, the tailpipe discharge momentum and vehicle-induced turbulence dominate the initial, rapid plume dispersion. Afterward, in the far-field regime, the plume dispersion is driven by the prevailing wind (Chan et al., 2001). In view of intermittency, the sampling duration ∆*τ* should be long enough to capture representative statistical properties. The current recommended sampling 84 duration for plume chasing is at least 2 minutes ($\Delta \tau \geq 352h/U_{\infty}$ where *h* and U_{∞} are the characteristic size and speed of the vehicle, respectively; Wang et al., 2020), which, however, is hardly realizable. Some of the emission parameters, such as engine power and vehicle speeds, are seldom constant. Moreover, the vehicle pair must travel a long distance together for one single test that arouses logistic concern. Apparently, a shorter sampling duration for plume chasing (but reliable readings) would be beneficial. Whereas, there is no study available for the drawback especially the sampling inaccuracy. The uncertainty of plume chasing in response to shortening sampling duration is analysed in this paper to bridge the knowledge gap.

 The data sensitivity to sampling duration in long-term, ambient air pollutant measurements has been studied for years. In annual averaging, it was mainly caused by synoptic scales or seasonal factors but not intermittency nor turbulent wakes (Brown and Woods 2014). On top of equipment precision, the measurement accuracy depends on sampling 97 duration $\Delta \tau$ and (unsteady) concentrations ϕ (Ballesta 2005, Brown et al. 2008). Venkatram (2002) examined the effect of sampling duration based on a binomial model of pollutant concentrations. However, the setting was oversimplified that barely represented the real-world situation. The power law

$$
\frac{\overline{\phi}_{\text{max}}}{\overline{\Phi}} = \left(\frac{\Delta \tau}{\Delta T}\right)^{-p} \tag{1}
$$

101 was suggested to describe the dependence of maximum mean concentration ϕ_{max} on (a shorter) 102 sampling duration $\Delta \tau$ (peak-to-mean ratio; Santos 2019). Here, $\overline{\Phi}$ is the mean concentration 103 over a longer sampling duration ΔT and p (a real number between 0 and 1) is the exponent 104 (Singer 1963). Theoretically, ΔT is long enough for asymptotically converged $\overline{\Phi}$ though it is hardly defined in non-stationary turbulence (Santos et al. 2009). Nonetheless, it is practically 106 employed as the reference to estimate the maximum mean concentration ϕ_{max} based on a 107 shorter sampling duration $\Delta \tau$ (Wilson 2010). However, the aforementioned studies have focused on (short-term) maximum but not the uncertainty induced by finite sampling duration. Apart from gaseous pollutants, similar findings have been arrived based on the transport of aeolian sediment (Ellis et al., 2012; Webb et al., 2019). Previous studies related to vehicular exhaust, by and large, have focused on the uncertainty of emission factor (EF; pollutant-to- carbon-dioxide concentration ratio) rather than mean concentration (Tong et al., 2022; Wang et al., 2020; Zheng et al., 2016). EF could avoid temporal variance and turbulence interference, however, its validity is based on stoichiometric combustion. While it is hard to maintain on- road complete combustion at all times, instrumentation issues, such as slow response time and/or short sampling duration, occur very often (Park et al. 2011), deviating from the above assumption. Besides, the determination of EF for individual pollutants from tailpipe emission depends on the reliable pollutant measurements in both plume and ambient (Wen et al., 2019). These uncertainties would degrade the quality of remote sensing. Under this circumstance, statistically robust measures of mean pollutant concentrations in plume chasing are alternative solutions.

122 Given a time trace of length *T*, the ideal sampling duration $\Delta \tau$ should be long enough 123 such that the mean concentration ϕ is independent (quasi-steady state). Under homogeneous and stationery turbulence, this independence is achievable provided that the sampling duration is longer than the time scale of turbulence eddies (Santos et al., 2009). In the light of the intermittent vehicular wake in plume chasing, a proper sampling duration is hardly defined. 127 Therefore, it is necessary to quantify the uncertainty of mean-concentration measurements induced by the sampling duration in plume chasing. Large-eddy simulation (LES) is adopted in this paper so the influence other than vehicular wake is excluded.

 This paper is organized as follows. Section 1 (this section) is the introduction. Next, the LES setup and the statistical methods for sampling uncertainty are described in Section 2. In 133 Section 3, the fluctuating concentrations behind the tailpipe within the near wake are analysed. Their power spectra are then employed to elucidate the relationship between uncertainty and sampling duration. Finally, the findings and the conclusions are summarized in Section 4.

2. Methodology

2.1 Mathematical Model

 LES is an appealing tool investigating the spatio-temporal dynamics of flows and pollutant dispersion (eddy-resolving). It explicitly calculates much of the conservation of momentum and mass while models small portion of subgrid-scale (SGS) fluxes at a reasonable computational load (Chan et al. 2008). LES is advantageous in terms of calculating the unsteadiness and intermittency of flows and tailpipe dispersion (Li et al. 2007) so is adopted.

 The LES used in this paper is the open-source computational fluid dynamics (CFD) code OpenFOAM 6 (Weller et al. 1998). It was validated in our previous study (Xie et al. 2020). The SGS motions are modelled by the Smagorinsky model (Smagorinsky 1963). The 147 computational domain sizes $31.8h$ (streamwise) \times 3.9*h* (spanwise) \times 10.3*h* (height) while the 148 model of the heavy-duty vehicle sizes 3.86*h* (length) \times 0.89*h* (width) \times 1.09*h* (height). The logarithmic law-of-the-wall (log-law) is used to model the flows near all the solid boundaries (the ground and the truck body). At the domain top and the spanwise extent, Neumann 151 boundary conditions (BCs; $\partial \psi / \partial n = 0$ where *n* is the normal to the boundary) for both flows 152 and pollutant transport are applied. Dirichlet BCs of constant wind speed U_{∞} and zero pollutant $\phi = 0$ are prescribed at the inflow. Turbulence is not prescribed at the inflow but is only induced by the flows around the vehicle. This configuration helps focus on tailpipe dispersion driven by wake-induced turbulence. An open BC is adopted at the outflow so all the pollutants are removed from the computational domain without any rebound. A point source of pollutant with 157 a constant emission rate Q is placed at the tailpipe exit $(x = y = z = 0)$ to simulate vehicular exhaust. Here, *x*, *y* and *z* are the streamwise, spanwise and vertical coordinates, respectively. The spatial domain is discretized into 3.38 million unstructured hexahedra using the mesh generation utility *snappyHexMesh* (OpenFOAM 2018). Its mesh is refined toward the vehicle 161 surfaces and the ground. The minimum and maximum cell volume is about $10^{-7}h^3$ and $10^{-2}h^3$, respectively. The second-order-accurate finite volume method (FVM) is used to discretize the 163 gradient, divergence and Laplacian terms. The time increment is $\Delta t = 0.15h/U_{\infty}$ and the LES is 164 integrated for $T = 510h/U_{\infty}$ in the time domain using the implicit, second-order-accurate backward differencing.

Fig. 1 (a) Digital model of the heavy-duty vehicle together with (b) computational domain and boundary conditions.

2.2 Statistical Method

 The gaseous pollutant considered in this paper is passive and chemically inert that could 169 be taken as carbon dioxide $CO₂$. It is diluted by vehicle-induced turbulence in the wake with characteristic scales of length *h* and velocity *U*∞. In favour of detection sensitivity, sampling within the near wake is suggested where the concentrations are almost ten times larger than those in the far field (Xie et al. 2020). The sampling locations are aligned along the sampling line in the streamwise direction at the tailpipe level from (0.05*h*, 0, 0) to (*h*, 0, 0) to mimic plume chasing (Huang et al., 2020b). The definition of variables used in this paper is summarized in Table. S1.

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179 **2.2.1 Moving Average**

180 The concentrations ϕ are normalized by the characteristic pollutant concentration Φ_0 (= 181 $\dot{Q}/U_{\infty}h^2$). Time traces of dimensionless pollutant concentration C_i (= ϕ_i/Φ_0) are probed from 182 the LES dataset where the subscript *i* is the index of the data sample. Their moving average

$$
\overline{C}\left(\Delta\tau\right)_i = \frac{1}{n} \sum_{j=0}^{n-1} C_{i+j} \tag{2}
$$

183 represents the sample mean over the sampling duration $\Delta \tau$ (= $(n-1)\times \Delta t$, where $n > 1$) is the 184 number of data points within $\Delta \tau$). Here, the overbar φ denotes time average. The sampling 185 duration considered is in the range of $10\Delta t \leq \Delta \tau \leq 320\Delta t$. It is noteworthy that 186 $\overline{C}_{i=0} (\Delta \tau = T = 510 h/U_{\infty})$ is the population mean \overline{C} .

187

188 **2.2.2 Uncertainty Analysis**

189 The relative deviation between the mean of each data subset (moving average) with 190 sampling duration $\Delta \tau$ and the population is

$$
\delta(\Delta \tau)_i = \frac{\overline{C}(\Delta \tau)_i - \overline{C}}{\overline{C}}.
$$
\n(3)

191 To consolidate the uncertainty induced by the data subsets with sampling duration ∆*τ* the 192 coefficient of variance

variance
\n
$$
CV(\Delta \tau) = \left\{ \frac{1}{N} \sum_{i=0}^{N-1} \left[\delta(\Delta \tau)_i \right]^2 \right\}^{1/2} = \left\{ \frac{1}{N} \sum_{i=0}^{N-1} \left[\frac{\overline{C}(\Delta \tau)_i - \overline{C}}{\overline{C}} \right]^2 \right\}^{1/2}
$$
\n(4)

193 is adopted where *N* is the number of data subsets with sample mean $C(\Delta \tau)$ *i*. It is noteworthy 194 that $CV(\Delta \tau = \Delta t)$ is equal to the fluctuating concentration intensity *I*.

For demonstration purposes, the sample mean $C(\Delta \tau)$ is defined acceptable in this 195 196 paper provided that its tolerance is within $\pm 15\%$ compared with the population mean C, i.e. 197 $|\delta(\Delta \tau)_i| \le 15\%$. To obtain a sample mean with specified confidence, the sampling duration $\Delta \tau$ 198 should be long enough so that more than 90% of the data in a new dataset $C(\Delta \tau)$ *i* are within 199 the acceptable deviation. Therefore, the fraction of acceptable sample mean

$$
k(\Delta \tau) = \frac{\text{No. of data subsets in which } |\delta(\Delta \tau)| \le 15\%}{N}
$$
 (5)

200 is studied. The specific criterion aforementioned was adopted elsewhere (Li et al. 2017).

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²⁰² **3. Results and Discussion**

203 **3.1 Fluctuating Concentration**

 Fig. 2a shows the population mean *C* , maximum *Cmax* and minimum *Cmin* of dimensionless pollutant concentration based on the entire LES dataset of sampling duration *T*. 206 Liu et al. (2011) found that a dimensionless averaging time T^* (= $T \times U_{ref}/L_{ref}$ where U_{ref} and *L_{ref}* are the reference scales of wind speed and length, respectively) in the range of 200 to 400 is sufficient for reliable population mean *C* around a high-rise building. The current 209 dimensionless averaging time $(T^* = T \times U_{\infty}/h = 510)$ well exceeds the requirement.

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 The initial dilution directly behind the tailpipe is dominated by the jet-like flows from the truck underbody. Simultaneously, the spanwise and vertical dispersion is attributed to the turbulence and instability generated by the shear within the jet-like flows (Xie et al. 2020). In 214 this connection, the population mean concentration C decreases monotonically for $x \le 0.6h$ then keeps at a low level thereafter (Fig. 2a). For example, the population mean concentration

Xie et al. (2022)

 C at $x = 0.4h$ is almost 14 times larger than that at $x = 0.8h$. The region close to $x = 0.8h$ is 217 characterized by the strong entrainment and the upward flows within the major recirculation, driving the jet-like flows back to the vehicle body (Fig. S1a and S1b). A similar sharp drop in concentration (an order of magnitude) after a vehicle was also reported by Chang (2009).

222 Fig. 2 Longitudinal profiles of (a) population mean C, maximum C_{max} and minimum C_{min} of dimensionless pollutant concentration together with (b) fluctuating concentration intensity *I* along the sampling line.

 In addition to the upward flows within the major recirculation, the fresh-air entrainment from the sides of the vehicle quickly dilutes the pollutant (Fig. S1a). It is concurred by the 228 close-to-zero minimum concentration C_{min} for $0.6h \le x \le h$ (Fig. 2a). Likewise, the maximum 229 concentration C_{max} elevates for $x \ge 0.6h$ though the population mean concentration C keeps decreasing behind the tailpipe (Fig. 2a). For example, the maximum concentration *Cmax* is 231 almost 6 times larger than the population mean C at $x = 0.8h$. It is in turn suggested that the 232 pollutant dispersion behind $x = 0.6h$ is intermittent in response to the strong shear-generated turbulence within the major recirculation.

 The fluctuating concentration intensity increases gradually in the streamwise direction 235 for $x \le 0.6h$ then soars thereafter, resulting in an elevated level ($I \ge 1$) towards the end of the near-wake region (Fig. 2b). The peaked *I* is over unity so the fluctuations are comparable to the mean *C* . The initial increase in *I* is attributed to the recirculating flows. Moreover, the larger eddies augment turbulent mixing (widening plume coverage). A similar plume development with increasing *I* in the near-source region over open terrain was reported 240 elsewhere (Yee and Biltoft, 2004). In the region $0.8h \le x \le h$, the flow entrainment and the shear in-between the near and far wakes contribute much to the elevated fluctuating concentrations.

245 Fig. 3 (a) Probability density function (PDF) of the relative deviations δ_i of instantaneous 246 pollutant concentrations at the sampling points $x = 0.2h$, 0.5*h* and 0.8*h* directly behind 247 the tailpipe. (b) Skewness and kurtosis of δ_i along the sampling line.

249 The probability density functions (PDFs) of relative deviation $\delta(\Delta \tau)_i$ at $x = 0.2h$, 0.5*h* and 0.8*h* are depicted in Fig. 3a. In view of the gradually augmented fluctuating concentration, the range of the PDF spreads with increasing distance behind the truck. Besides, the PDF of

 $\delta(\Delta \tau)$ at $x = 0.2h$ and 0.5*h* is close to Gaussian distribution but at $x = 0.8h$ is positively skewed. 253 As such, most of the measured instantaneous concentrations at $x = 0.8h$ are lower than the population mean *C* . The asymmetric PDFs are concurred by the skewness and kurtosis of $\delta(\Delta \tau)_i$ which are close to zero and 3, respectively, for $x \leq 0.6h$ (Fig. 3b). Thereafter, the increasing skewness and kurtosis indicate the positively skewed and leptokurtic PDFs. The 257 sharp change in maximum relative deviation δ_{max} from $x = 0.5h$ to $x = 0.8h$ is also notable (Fig. 3a). It is attributed to the turbulence generated by the major recirculation and the entrainment. 259 Likewise, the minor increase in maximum δ_{max} (the upper range of δ_i shown in Fig. 3a) for 0.2*h* $\leq x \leq 0.5h$ is attributed to the plume development driven by the recirculation.

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262 **3.2 Uncertainty Analysis**

The mean concentrations at different sampling duration $C(\Delta \tau)$ _i are calculated by 263 264 moving average based on the instantaneous (dimensionless) concentration *Ci*. The averaging 265 time $\Delta \tau$, which is also the sampling duration, cannot be too long, owing to the finite total 266 sampling period *T*. Otherwise, the data are insufficient for the analysis of sample mean, leading 267 to substantial inaccuracy. The sampling duration is limited to $\Delta \tau \leq 320 \Delta t$ that is roughly 9% of 268 the total sampling period *T*. It also satisfies the requirement of averaging time $\Delta \tau$ that is less 269 than one-third of the entire time trace *T* (Janik et al, 2012). Fig. 4 compares the time traces of 270 the relative deviations $\delta(\Delta \tau)$ *i* for different sampling duration at $x = 0.8h$. It is found that the 271 instantaneous relative deviations δ_i are highly fluctuating whose maximum is up to $\delta_{max} = 650\%$. 272 It is in turn implied that the maximum concentration *Cmax* is up to 6.5 times larger than the 273 population mean *C*. The variation of the relative deviations for sample mean $\delta(\Delta \tau > \Delta t)_i$ is less 274 than that of the instantaneous value $\delta(\Delta \tau = \Delta t)_i$. Increasing the sampling duration $\Delta \tau$ reduces 275 the uncertainty of the sample mean concentrations. For example, the maximum relative 276 deviation $\delta(\Delta \tau)_{max}$ for $\Delta \tau = 160 \Delta t$ is only 60%. The improved accuracy is attributed to the low

pass of sample mean $C(\Delta \tau)$, with period shorter than the averaging time (sampling duration) 277 $\Delta \tau$ by applying moving average. The average of sample mean concentrations $C(\Delta \tau)$ _i obtained 278 279 by moving average is very close to the population mean *C* . However, parts of the fluctuating 280 signal, especially those short-term extremities, are filtered out (Fig. 4).

282

283 Fig. 4 Time traces of the relative deviation $\delta(\Delta \tau)_i$ of pollutant concentrations. Sampling 284 duration $\Delta \tau = (a) \Delta t$ (instantaneous values); (b) $10\Delta t$; (c) $40\Delta t$; and (d) $160\Delta t$ by moving 285 average behind the tailpipe at $(x, y, z) = (0.8h, 0, 0)$.

286 Cumulative density functions (CDFs) measure the coefficient of variation *CV*(Δ*τ*) by 287 the slope of the curve core (Fig. 5). Steeper gradient suggests a smaller coefficient of variation, 288 and vice versa. Moreover, it depicts the maximum relative deviation $\delta(\Delta \tau)_{max}$ (along the *x*-axis 289 of Fig. 5) at which the CDF reaches unity (Santos et al., 2005). Like Fig. 3, the CDF (Fig. 5) 290 shows that the relative deviation for $\Delta \tau = \Delta t$ at $x = 0.8h$ is positively skewed, signifying frequent 291 low-concentration and occasional high-concentration events. The sampling duration Δ*τ* has a 292 strong influence on the maximum relative deviation $\delta(\Delta \tau)_{max}$ which decreases by almost ten 293 times when the sampling duration is increased from $\Delta \tau = \Delta t$ to $\Delta \tau = 160 \Delta t$. As shown in Fig. 294 5, the curve core steepens with extending sampling duration $\Delta \tau$, indicating a smaller coefficient of variation $CV(\Delta \tau)$ as well as more accurate sample mean concentrations $C(\Delta \tau)$ _i. It is noticed 295 296 that, with increasing sampling duration $\Delta \tau$, the CDF gradually converges close to normal 297 distribution that is in line with that reported elsewhere (Venkatram 2002).

299 Fig. 5 Cumulative density functions (CDFs) of the relative deviation $\delta(\Delta \tau)$ *i* of instantaneous 300 concentrations ($\Delta \tau = \Delta t$) and the sample mean concentrations averaged over $\Delta \tau = 10 \Delta t$, 301 $40\Delta t$ and $160\Delta t$ at $(x, y, z) = (0.8h, 0, 0)$. Dashed lines denote the CDFs of corresponding 302 normal distribution with the same mean and standard deviation.

306 Fig. 6 Relative deviations $\delta(\Delta \tau)$ *i* of sample mean concentrations obtained by moving average 307 over different sampling duration Δ*τ* at *x* = (a) 0.2*h*; (b) 0.5*h*; and (c) 0.8*h*. Also shown 308 are the frequency distribution of the absolute relative deviation $|\delta(\Delta \tau)_i|$ at $x = (d)$ 0.2*h*; 309 (e) 0.5*h*; and (f) 0.8*h* with $\Delta \tau = \Delta t$, 10 Δt , 40 Δt , and 160 Δt .

310 In Fig. 6a ($x = 0.2h$) and 6b ($x = 0.5h$), the relative deviation $\delta(\Delta \tau)$ *i* is almost normally 311 distributed considering its symmetry about zero (Fig. 3a). As shown in Fig. 6c, the positive tail 312 diminishes with increasing sampling duration $\Delta \tau$ so the relative deviation $\delta(\Delta \tau)_i$ at $x = 0.8h$ 313 tends to be normally distributed. It is also found that increasing the sampling duration $\Delta \tau$ narrows the range of the sample mean concentrations $C(\Delta \tau)$. On the contrary, the extreme 314 315 sample mean concentration $C(\Delta \tau)_{max}$ rises sharply with shortening sampling duration.

316

317 Figs. 6d, 6e and 6f depict the frequency distribution of the absolute relative deviation 318 $|\delta(\Delta \tau)_i|$ at the three sampling locations. As shown in Fig. 2a, the fluctuating concentration 319 intensity *I* at $x = 0.2h$ and 0.5*h* is lower than that at $x = 0.8h$. It is thus implied that at $x = 0.2h$ 320 and 0.5*h* (Fig. 6d, 6e), the fraction of the data close to the population mean C, such as $|\delta(\Delta \tau)_i|$ 321 \leq 10%, is much higher than that at $x = 0.8h$ (Fig. 6f) for the same sampling duration $\Delta \tau$. Taking 322 the data subset with sampling duration $\Delta \tau = 160 \Delta t$ as an example, almost 90% and 60% of the 323 absolute relative deviation $|\delta(\Delta \tau)|$ are less than 10% at $x = 0.2h$ and 0.5*h*, respectively. On the 324 contrary, only 20% of $|\delta(\Delta \tau)| \le 10\%$ at $x = 0.8h$ where the fluctuating concentration intensity 325 *I* is much larger. However, the fraction of instantaneous dimensionless concentrations 326 $C(\Delta \tau = \Delta t)$ within the same range is only 50% at $x = 0.2h$, 20% at $x = 0.5h$ and 10% at $x =$ 327 0.8*h*. It is hence suggested that for the region close to the tailpipe with low fluctuating 328 concentration intensity *I*, more accurate sampling is achievable using a shorter sampling 329 duration $\Delta \tau$. The difference in the sampling accuracy $|\delta(\Delta \tau)|$ between $x = 0.2h$ and $x = 0.8h$ 330 reduces after applying a longer sampling duration $\Delta \tau$. It is because more short-term extremities 331 are filtered out.

333 Fig. 7 Coefficient of variance $CV(\Delta \tau)$ for instantaneous concentration ($\Delta \tau = \Delta t$) and sample 334 mean with (a) sampling duration $\Delta \tau = 10 \Delta t$; 40 Δt ; and 160 Δt along the sampling line 335 together with (b) different sampling duration $\Delta \tau$ at the selected sampling locations.

337 It is observed that increasing the sampling duration $\Delta \tau$ reduces the uncertainty (Fig. 7a) 338 in the entire major recirculation. The improvement is more obvious in the range of $0.6h \leq x \leq$ 339 *h* where the concentrations are highly fluctuating in response to the underbody flows and 340 sideward entrainment (Fig. S1). Although the sampling duration is extended to $\Delta \tau = 160 \Delta t$, the 341 uncertainty in sample mean concentration for $x \ge 0.6h$ soars. Such a phenomenon is attributed 342 to the instantaneous fluctuating dimensionless concentration C_i - C in $0.6h \le x \le h$ that is 343 tightly driven by the eddies in the major recirculation. Their effect is not negligible unless the 344 sampling duration $\Delta \tau$ is longer than the turbulence time scales. The coefficient of variance 345 *CV*($\Delta \tau$) decreases with increasing sampling duration $\Delta \tau$ (Fig. 7b). Its diminishing gradient 346 indicates the importance of sampling duration $\Delta \tau$ to accuracy. Given a sufficiently long $\Delta \tau \geq 1$ 347 160Δ*t*, the curves flatten so the sampling uncertainty is negligible. Further increasing the 348 sampling duration $\Delta \tau$, however, leads to costly measurement but limited accuracy improvement.

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350 Fig. 8 Fraction $k(\Delta \tau)$ of instantaneous concentrations and sample mean concentrations whose 351 relative deviations are within $\pm 15\%$ ($|\delta(\Delta \tau)| \le 15\%$). (a) Along the sampling line behind 352 the tailpipe and (b) with different sampling duration $\Delta\tau$ at the sampling locations.

354 A key question is how long the sampling duration $\Delta \tau$ is sufficient for reliable plume chasing. For demonstration purposes, the range of reliable sample mean concentration $C(\Delta \tau)$ 355 356 is herewith defined as $\pm 15\%$ of the population mean C, i.e. $|\delta(\Delta \tau)_i| \le 15\%$. The fraction $k(\Delta \tau)$ 357 of data sample within this range is compared in Fig. 8. The minimum sampling duration 358 enabling a reliable sample mean is defined as the shortest averaging time under which $k(\Delta \tau) \ge$ 359 90% (Li et al. 2017). The fraction $k(\Delta \tau)$ for different sampling duration $\Delta \tau$ decreases in the 360 streamwise direction until $x = 0.6h$ that remains at a low level thereafter (Fig. 8a). When 361 adopting the (longer) sampling duration of $\Delta \tau = 160 \Delta t$, the data subset for sampling locations 362 $x \le 0.4h$ fulfils the 90% criteria but not for $x \ge 0.6h$ where $k(\Delta \tau)$ is only about 40%. Therefore, 363 plume chasing targeting within $x \leq 0.6h$ enables more accurate measurements for the same 364 sampling duration $\Delta \tau$. Indeed, the uncertainty could be further reduced if the sampling points 365 are closer to the tailpipe. The acceptable sampling duration $\Delta \tau$ is shortened to 40 Δt at $x = 0.2h$ 366 that is reduced by 5 times compared with $220\Delta t$ at $x = 0.5h$ (Fig. 8b). Whereas, the 90%

367 criterion is not achievable at $x = 0.8h$ for the range of sampling duration $\Delta \tau$ tested. Unlike the 368 other two sampling points, the dispersion at $x = 0.8h$ is more affected by energetic eddies whose influence is hardly eliminated by averaging over a finite sampling duration. It is noteworthy 370 that the sampling duration in the current sensitivity test is limited to $\Delta \tau \leq 320 \Delta t$ to ensure validity (Fig. 8b). Otherwise, the number of sample mean would be insufficient, degrading the subsequent error analysis of sample mean. Similar concern was reported elsewhere (Janik et al. 2012).

3.3 Fast Fourier Transform

 Fast Fourier Transform (FFT) is adopted in this study to investigate the frequency characteristics of the tailpipe dispersion within the near wake. It transforms the data from time domain to frequency domain, providing the power associated with different frequencies (Richards 2003). Fig. 9 shows the power spectra of relative deviations of instantaneous 380 concentrations $\delta(\Delta \tau)_i$ at the three sampling locations. The frequency is normalized in the form 381 of Strouhal number $St = fd/U_{\infty}$ where *f* is the frequency and *d* the trunk width (McArthur et al. 2016). The power generally increases with increasing frequency in the low-frequency regime, 383 reaches its maximum in $0.03 \leq St \leq 0.1$ and decreases thereafter. It finally keeps at a low level 384 ($\leq 10^{-3}$) for $St \geq 1$. The unsteadiness in concentration is directly affected by the flow intermittency. Therefore, the spectra obtained from the concentration data help identify the dominant scales in the near-wake region.

Fig. 9 Power spectra of relative deviation $\delta(\Delta \tau = \Delta t)_i$ for instantaneous concentrations at the selected sampling locations at $x = (a) 0.2h$; (b) 0.5*h*; and (c) 0.8*h*. Also shown in (c) are the power spectra of relative deviations $\delta(\Delta \tau = 40\Delta t)$ for sample mean concentrations over $\Delta \tau = 40\Delta t$ obtained by moving average and the corresponding sample frequency $f = 1/\Delta \tau = 1/40\Delta t$ (dark solid line). The primary and secondary peaks for $\Delta \tau = \Delta t$ are highlighted (circles in (c)).

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390 The pollutant transport for $x \le 0.6h$ is mainly driven by the jet-like flows from the 391 vehicle underbody (Fig. S1a). Therefore, the low-frequency motions, which is peaked at $St =$ 392 0.033, are more energetic (Fig. 9a and 9b). At *x* = 0.8*h*, a primary peak and a secondary peak 393 are shown at $St = 0.084$ and $St = 0.19$, respectively (Fig. 9c). Alike Volpe et al. (2015), the two 394 peaks are attributed to the wake pumping of major recirculation $(St = 0.08)$ and the vortex 395 shedding initiated at the two vertical edges of the vehicle $(St = 0.19)$. Wake pumping is the key 396 component in the unsteady recirculation. It is induced by the wake lengthening and shortening 397 in response to the increasing entrainment into the near wake together with the vortex shedding 398 induced by the major recirculation (Richards 2003, Rao et al. 2019).

399

400 The power spectra of relative deviation of the instantaneous concentrations δ_i and the 401 sample mean concentrations $\delta(\Delta \tau = 40\Delta t)$ *i* are compared to examine the effect of moving 402 average in the frequency domain (Fig. 9c). In fact, moving average applies a low-pass filter on 403 the data in the time domain. It damps out the signal with frequency higher than $1/\Delta\tau$ (cut-off 404 frequency). The power of low-frequency signal is less affected. The power spectral density 405 (PSD) of sample mean obtained by moving average decreases by a factor of $\sin^2(\pi f \Delta \tau)/(\pi f \Delta \tau)$ 406 compared with the original ones. The difference diminishes when the product of frequency *f* 407 and duration $\Delta \tau$ is larger than unity (Arya, S. P., 1999). As shown in Fig. 9c, the sampling 408 duration, which can capture the dominant frequency at *x* = 0.8*h*, should be higher than 70Δ*t*. 409 Such sampling duration is roughly the transition point for $CV(\Delta \tau)$ to stabilize with increasing 410 $\Delta \tau$ (Fig. 7). More high-frequency signals could be averaged out with longer $\Delta \tau$. If $\Delta \tau$ is long 411 enough to capture the dominant frequency (signals with most energy), the improvement in 412 measurement accuracy would be slowed down. The results suggest that a short sampling

 duration Δ*τ* only captures the high-frequency signal. Sampling signal of dominant frequency is crucial to the measurement accuracy of mean concentrations.

 It is known that the spectra of fluctuating concentration depend on the distance from the point source (Mylne and Mason 1991). In the vicinity of a tailpipe, the turbulence characteristic length scale (wake-induced) is usually longer than the plume width (Xie et al. 2007), resulting in plume meandering. Eddies dominate the transport as long as the plume coverage is comparable to or larger than the turbulence characteristic length scale. Trunk dimension is the characteristic length scale in the near-wake region after a heavy-duty vehicle. The plume transport is mainly driven by the vehicular wake, especially the wake pumping and the vortex shedding from the longer trunk edges.

3.4 Implication to plume chasing

 The results reported above collectively show that, in plume chasing deployment, increasing the sampling duration helps filter out parts of fluctuating signal as well as reduce sampling uncertainty. The sample mean often fluctuates substantially for a short sampling duration. As such, it would possibly deviate much from the population mean. The sample mean varies less with extending sampling duration. Therefore, a longer sampling duration is more favourable for a reliable sample mean as well as the tailpipe emission. As shown in the frequency analysis, the sampling accuracy could be affected by the dominant frequency. If the sampling duration is long enough to capture the signal at the dominant frequency (inverse of sampling duration smaller than the dominant frequency), the sampling accuracy would be improved substantially. An even longer sampling duration, which is longer than the inverse of the dominant frequency, however, would slow down the improvement in sampling accuracy.

437 The threshold sampling duration is defined as the shortest time period over which the fraction 438 of data sample satisfying $|\delta(\Delta \tau)_i| \le 15\%$ reaches 90%. The threshold sampling durations are 439 40 Δt and 220 Δt , respectively, at $x = 0.2h$ and $x = 0.5h$ after the truck. However, it is much 440 longer at $x = 0.8h$ that is beyond the range of sampling duration being investigated in this paper. 441 For a 4-m-high truck driving at a speed of 10 m sec⁻¹, the sampling duration should be at least 442 2.4 sec at $x = 0.8$ m or 13.2 sec at $x = 2$ m. A shorter sampling duration is required to obtain a 443 reliable sample mean in the region close to the tailpipe. In view of the elevated fluctuating 444 concentration intensity in the region after $x = 0.6h$, a longer sampling duration is needed. 445 Therefore, in the plume chasing after a heavy-duty vehicle, it is suggested to sample within the 446 region $x \le 0.6h$. Moreover, the measurements would be more reliable if the sampling point is 447 closer to the tailpipe.

448

⁴⁴⁹ **4. Conclusions**

450

451 In order to investigate the effect of sampling duration $\Delta \tau$ on vehicular pollutant measurement, LES is carried out for a heavy-duty vehicle to collect the spatio-temporal behaviours of pollutant concentrations at the tailpipe level within the near-wake region. The sampling uncertainty is then examined by statistical analysis. Based on the results reported above, the conclusions could be drawn as follows.

456

457 Within the near-wake region, the fluctuating concentration intensity *I* increases slowly for 458 $x \leq 0.6h$. It experiences a sharp increase thereafter due to the augmentation of fluctuating 459 concentration by the major recirculation. Afterward, a positively skewed distribution after

460 $x = 0.6h$ is developed, indicating that the instantaneous concentrations could have a notable 461 deviation from the population mean *C* .

462 A longer sampling duration would result in the loss of the high-frequency fluctuating 463 components, leaving the low-frequency signal. Thus, the coefficient of concentration 464 variance $CV(\Delta \tau)$ would decrease with increasing sampling duration (i.e. more accurate 465 sample mean). However, the improvement in sampling accuracy gradually diminishes if 466 the sampling duration is longer than $160\Delta t$. It is noteworthy that, even a long sampling 467 duration is adopted, the sampling accuracy degrades for $x \ge 0.6h$ because of the elevated 468 fluctuating concentration intensity *I*.

469 • Sampling duration also affects the distribution of sample mean concentration $C(\Delta \tau)$ *i*. For 470 a longer sampling duration, the maximum sample mean $C(\Delta \tau)_{max}$ and the minimum C 471 $(\Delta \tau)_{min}$ approach the population mean C, improving the sampling accuracy. However, the 472 improvement lessens for prolonging sampling duration Δ*τ*. Increasing the sampling 473 duration helps the distribution of sample mean $C(\Delta \tau)_i$ that is alike the normal distribution 474 with increasing sampling duration.

475 • The minimum sampling durations are $40\Delta t$ at $x = 0.2h$ and $220\Delta t$ at $x = 0.5h$. However, at $x = 0.8h$, the minimum sampling duration is even longer than the entire time period *T* being collected in this study. A shorter sampling duration is needed to acquire a reliable sample mean concentration in the region close to the tailpipe.

479 From the FFT analysis, it is found that the variance of sample mean is attributed to the 480 signal with frequency lower than the sampling frequency $(= 1/\Delta \tau)$. This indicates that 481 sampling at the dominant frequencies could reduce the sampling uncertainty to a large 482 extent.

 The aforementioned findings collectively enrich our understanding of how the sampling 485 uncertainty of plume chasing varies with the sampling duration $\Delta \tau$ behind the tailpipe within the near-wake region. In this study, the variation of uncertainty is mainly attributed to the pollutant source and the turbulence of vehicular wake. In practice, vehicular emission in the wake region could also be affected by some other factors, such as engines, acceleration, and brakes. This paper only focuses on the turbulence effect induced by the vehicle body. Further studies could combine with the sampling uncertainty in field measurements to advance the contribution from different factors. Although it is advised to sample pollutant concentrations within the near-wake region, practically the safe distance apart should be at least 10 m (≥ 2*h* in this paper). In this connection, it is worthy to look into the sampling accuracy beyond the near wake.

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661 **Supplementary Materials**

662 Table. S1 Nomenclature.

664

665 Fig. S1 Shaded contours of dimensionless pollutant concentration C_i (= ϕ_i/Φ_0) overlaid with 666 streamlines on (a) *x*-*z* plane at the centreline ($y = 0$) and (b) *x*-*y* plane at the tailpipe 667 level $(z = 0)$.