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NOx and fine particles Yoshihiro Nakashima^{1*}, Charlotte E. Jones², Wakana Yamanobe³, Yoshizumi Kajii² ¹ Faculty of Agriculture, Tokyo University of Agriculture and Technology, 3-8-1 Harumi-cho, Fuchu, Tokyo, Japan ² Graduate School of Global Environmental Studies, Kyoto University, Yoshida-Honmachi, Sakyo-ku, Kyoto, Japan ³ Aprica Children's Products Japan Co. Ltd, 2-6-12 Ginza, Chuo-ku, Tokyo, Japan Abstract This paper presents daytime vertical profiles of the NOx concentration, and the number concentration and size distribution of fine particles near a major road in urban Tokyo during spring 2011. No significant height dependence was observed in the NOx concentration, presumably due to rapid diffusion. In contrast, the number concentration of particles under 0.5 µm diameter demonstrated an exponential decrease with increasing height above ground level. Vertical profiles derived from this study differ from those presented in previous studies, however, these differences may potentially be explained by the different tailpipe positions of Japanese and US heavy vehicles, as well as the meteorological conditions. This study demonstrates that in Tokyo, the fine particle concentration at 0.5 m above ground level was about 2.9 times higher than that at 2.0 m. The higher fine particle concentration immediately above ground level implies that children may be at greater risk of experiencing pollutant-related respiratory symptoms than

Near-surface vertical profiles of urban roadside

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adults.

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28 *Keywords:* Vertical concentration profile; High density traffic road in urban area; Fine particle.

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- 31 **INTRODUCTION**
- 32

33 Exhaust from heavy vehicles contributes one of the major anthropogenic source of the 34 gaseous pollutants CO, NOx and NMHCs, as well as fine particles. Fine particles in particular are 35 known to cause adverse health effects on humans and a relationship between exposure to high concentrations of fine particles and respiratory morbidity has been suggested (Li et al., 2008; 36 37 Peden 2005; Riedl 2008). Recently, an immune response has been observed in humans following 38 exposure to ultrafine particles with a diameter of less than 0.1 µm, (Li et al., 2009, 2010). Several epidemiological studies into respiratory symptoms caused by inhaling anthropogenic fine 39 40 particles have demonstrated that the prevalence of asthma may be related to regional motor 41 vehicle traffic density and residential proximity to freeways (Holguin 2008; Patel and Miller 42 2009; Salam et al., 2008), while other studies reported that there was no evidence to suggest such 43 a relationship (Waldron et al., 1995; Livingstone et al., 1996). To evaluate the correlation 44 between vehicular emissions of fine particles and any potential health impacts, detailed 45 measurements of the concentration and spatial distribution of fine and ultrafine particles are essential. 46

The spatial distribution of fine particles has been reported for highway and freeway locations
(Zhu *et al.*, 2002; Zhu and Hinds 2005; Zhu *et al.*, 2006; Pohjola *et al.*, 2007; Hagler *et al.*, 2009;
Hu *et al.*, 2009; He and Dhaniyala 2012). It has been reported that the horizontal distribution of
particle number concentration shows an exponential decay with increasing distance from a

51	highway, in the absence of any obstructions (Zhu et al., 2002). In contrast, the vertical
52	distribution of particles does not indicate exponential decay with increasing distance from the
53	emission source, although the maximum concentration typically occurs close to ground level (<4
54	m). These studies were performed in areas surrounding a highway or freeway. However, it is
55	arguably more important to carry out these measurements in the vicinity of high traffic density
56	roads within urban areas.
57	This study focuses on determining the vertical profiles of the NOx concentration and number
58	concentration of fine particles within a busy urban street in central Tokyo. The concentrations of
59	NOx and fine particles were measured at heights from 0.5 to 2.0 m above ground level, since this
60	corresponds to the region where humans will be most directly affected by exposure to vehicular
61	emissions. In theory, young children and babies transported via a stroller may be the most
62	affected by air pollutants near the surface, due to both their relatively low height above ground
63	level and undeveloped respiratory system. The relationship between road traffic and adverse
64	respiratory effects in children has been studied previously (Weiland et al., 1994; Oosterlee et al.,
65	1996; Ciccone et al., 1998), and differences in the adverse respiratory effects of fine particles on
66	children and adults have been reported (Oosterlee et al., 1996)
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METHODS

70	Field measurements were conducted in urban Tokyo on April 4th, 2011. Fig. 1 shows the
71	location of the measurement site, which was near to the intersection of two main roads, oriented
72	in the direction of Northeast to Southwest (Route-15) and Northwest to Southeast (Route-304).
73	At a distance of about 300 m from the site, two local roads run parallel to Route-15 in the
74	direction of Northwest (Route-405) and Southeast (Route-3016), respectively. In addition, an
75	elevated highway surrounds the measurement site, at a distance of 300-600 m. The area is
76	relatively flat, with tall buildings (6-12 stories) located alongside all roads near to the site. Due to
77	the built up nature of the area, particle emissions from the road adjacent to the site were expected
78	to dominate the measurements, while contributions from other roads in the vicinity are thought to
79	be small. The measurements were carried out between 15:00-16:00 Japanese local time -
80	corresponding to a period of high road usage, with respect to both vehicles and pedestrians.
81	Traffic data for the local roads around the site was obtained from the Ministry of Land,
82	Infrastructure, Transport and Tourism. According to the data, the traffic volume for vehicles in
83	the daytime (7:00-19:00) is approximately 31,000 vehicles per day, with heavy duty vehicles
84	accounting for 16% of the total. The average vehicle speed on the road in typical, congested
85	conditions was 12 km h^{-1} .

86 The size distribution and number concentration of fine particles were measured using an 87 optical particle counter (OPC, model 1.109, Grimm). A laser diode was employed to detect and

88	count particulates. The size distribution was set from 0.25 to 3.0 μ m. Particles of less than 0.25
89	μ m in diameter cannot be detected by the instrument, since these particles are outside of the range
90	for detection. The OPC instrument continuously sampled air at 1.2 L min ⁻¹ and the concentration
91	of each particle size range was measured every 6 seconds. The air was supplied to the instrument
92	via PFA tube (6.35 mm outer diameter, 3 m length) and the inlet of the tube was fixed to a
93	moveable rod, such that the vertical height of the inlet from the ground could be changed
94	periodically. The inlet height was alternated between 0.25, 0.5, 1.0, 1.5 and 2.0 m above ground
95	level. The inlet and OPC were deployed about 1m away from the intersection. The accumulation
96	time was set to 2 minutes for each measurement height, with an interval of 1 minute following
97	each inlet height adjustment. The full vertical profile measurements were carried out 3 times and
98	the averaged particle number concentration was derived for each height. In addition to the
99	particle measurements, the vertical profile of nitrogen oxides (NO and NO ₂) was measured by O_3
100	chemiluminescence (Model 42i-TL, Thermo Electron) simultaneously via the same inlet system.
101	Unfortunately, meteorological data including ambient temperature, wind speed and direction
102	were not measured due to instrument difficulties. However, it is assumed that the difference of
103	the concentration of the trace species coming from upwind and downwind is small since the
104	roadside are surrounded by the tall buildings.

106 **RESULTS AND DISCUSSIONs**

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108 Vertical profiles of the concentrations of NO and NO₂ are shown in Fig. 2. No height 109 dependence was observed in the NO concentration, while a slight height dependence was 110 observed for NO₂, within the standard deviation of the measurement. Since gaseous species such 111 as NO and NO₂ can readily diffuse within the atmosphere, no strong vertical variation was 112 observed for NO and NO₂ within the range of sampling heights tested. The vertical profiles of 113 pollutant gases have been simulated for CO (Johnson et al., 1973). In scenarios where roadside 114 emission sources are surrounded by tall buildings, the so called "street canyon", emitted gases are 115 thought to be mixed by helical air circulation. The vertical profiles of pollutant gases for a height 116 z may be derived as follows;

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$$C(z) = C(0) \times \frac{x+2}{[(x^2+z^2)^{1/2}+2]}$$
(1)

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where *x* stands for the horizontal distance between the emission source and measurement point
(2.0 m in the present study), and *C*(0) is the surface concentration of the pollutant. Appling the Eq.
(1) to the present measurement, the ratio of the concentration between the heights of 0.25 and 2.0
m were 0.83. Since the standard deviation of the concentration for each sampling height is large,
no height dependence of the concentration simulated by Eq. (1) was observed clearly.

124 Fig. 3 shows the size distribution of fine particles for each sampling height. Whilst there was 125 virtually no significant height dependence observed in the NO and NO₂ mixing ratios, in contrast, 126 the number concentration of fine particles did demonstrate a degree of height dependence. For all 127 sampling heights, the maximum number concentration was observed for particles of size 0.25-128 $0.30 \,\mu\text{m}$, and the number concentration exponentially decreased with increasing particle size. Fig. 129 3 indicates that the particle size distribution is broadly uniform throughout all sampling heights, 130 suggesting that there is no unique formation or loss process of the particles occurring at any 131 individual sampling heights. However, the overall number concentration was seen to decrease 132 with increasing sampling height.

Fig. 4 shows the vertical profiles of the number concentration of particles for each size range. 133 134 Particles of diameter larger than 0.5 µm showed vertical concentration profiles which were almost uniform (within one standard deviation), while a clear height dependence was observed in 135 136 the concentration of the smaller particles. The concentration of particles within each size range decreased with increasing height, resulting in apparently exponential vertical profiles of the 137 138 number concentration. Vertical profiles of the number concentration of fine particles have previously been measured near a highway in September 2009 in Liverpool, New York (He and 139 140 Dhaniyala 2012) and close to a freeway in Los Angeles in July 2001 (Zhu and Hinds 2005). The 141 results from these studies indicate that the maximum particle concentration occurred at a height

142	of 3.4 m (He and Dhaniyala 2012) and 3.0 m (Zhu and Hinds 2005). He and Dhaniyala have
143	explained their results by the position of the tailpipe in heavy duty vehicles and the generation of
144	a thermal plume. Zhu and Hinds, on the other hands, have not explained for the measured vertical
145	profiles but there is the evidence that the ground level of measurement site was ~4.5 m lower than
146	the Freeway. In the present study, the tailpipe of heavy vehicles in Japan is located in almost
147	exactly the same position as that of normal commuter gasoline vehicles, ~0.5 m above ground
148	level. In addition, the measurements for this study were carried out in late afternoon in April, and
149	as such the ground was not significantly heated since the sunlight was blocked by the tall
150	buildings, which would reduce any thermal plume effect. Ground level of the sampling site is
151	almost same as that of the roadside. These differing conditions are consistent with the differences
152	in the vertical profiles of fine particles measured in this work and the previous studies. Since
153	different vertical profiles of roadside particulates have been observed in different locations,
154	further studies to determine the dependence of the particle number concentration and size
155	distribution on seasonal changes, as well as on the position of the tailpipe are necessary.
156	For each particle size, the ratio between the standard number concentration at each height and
157	the concentration at 2.0 m was calculated. For particles smaller than 0.5 μ m diameter, the ratio of
158	the number concentration increased exponentially as the sampling height decreased. The

159 maximum ratio was 2.9 for the 0.25-0.30 μ m size range. This indicates that the level of exposure

160 to fine particles, and therefore the potential for related adverse health effects, may be reduced 161 with increasing height above ground level. In light of this, it might be anticipated that children 162 would be more susceptible to health issues related to roadside fine particle levels compared to 163 adults. Health implications for children living near to areas with high traffic density have been 164 researched previously (Weiland et al., 1994; Oosterlee et al., 1996; Ciccone et al., 1998). 165 Ciccone *et al.* (1998), investigated chronic respiratory symptoms in children under fifteen years 166 of age and compared them with those of adults living in similar conditions. They concluded that 167 living in the vicinity of busy streets with high traffic flow increased the risk of developing 168 chronic respiratory symptoms in children, while there was no significant correlation in adults. Although they did not offer an explanation for the increased risk in children, fine particles are 169 170 thought to be a contributing factor.

Based on the results of this study, it is implied the importance of the reduction of the number concentration of fine particle at the lower height. It should be noted that ultrafine particles were not observed in this study, as ambient concentrations were below the instrument detection limit. Based upon the observed vertical profile of fine particles, it is probable that the same height dependence of the number concentration of ultrafine particles will be observed. Recently, a relationship between exposure to ultrafine particles and adverse health effects has been confirmed (Oberdörster *et al.*, 1994; Penttinen *et al.*, 2001; Li *et al.*, 2010). As such, in future studies to assess the impact of particulate emissions on human health, vertical profile measurements forboth fine and ultrafine particles would be desirable.

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181 CONCLUSIONS

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Vertical profile measurements of the number concentration of fine particles in an urban street 183 184 were carried out during the afternoon in spring 2011. The results obtained imply that the risk of 185 developing chronic respiratory symptoms may be greater for children compared to adults. The 186 exponential decay of the particle number concentration with increasing height above ground level 187 is thought to be the result of the low position of tailpipes on Japanese heavy vehicles and the 188 relatively cold road surface. To assess the risk of developing particulate-related respiratory 189 diseases as a result of exposure to vehicular emissions, both the horizontal and vertical 190 distribution of the fine particle concentration should be monitored. In summertime, strong 191 thermal plumes are created in urban areas due to the heat-island phenomenon. As such, it is 192 anticipated that the vertical profile will show seasonal variations. Thus the vertical profile 193 measurements should be repeated in different seasons, in order to achieve a more detailed 194 assessment of typical yearly exposure levels. In addition, in future studies, detailed measurements 195 including ultrafine particles, as well as fine particles should be carried out.

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- 267

269	Figure Captions
270	Fig. 1. An aerial photograph of the measurement site and the surrounding terrain.
271	Fig. 2. Vertical profiles of the concentration of NO (red) and NO ₂ (blue). Plots of NO ₂ are shifted
272	to 0.1 m higher in this figure. Error bars indicate one standard deviation.
273	Fig. 3. Size distribution of the fine particle for each sampling height.
274	Fig. 4. Vertical profiles of number concentration of fine particle for each particle diameter. Errors
275	bar indicate one standard deviation.











Fig. 2.







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Fig. 3.



Fig. 4.