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## <sup>23</sup> Characteristics of the pollutant emissions in a tunnel of Shanghai on a weekday

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#### ABSTRACT

Tunnel displays a typical semi-closed environment, and multitudes of the pollutants tend to accumulate. The samples of gaseous pollutants and particulate matter (PM) were 19 collected from the Xiangyin tunnel at Shanghai to investigate the characteristics of the 20 pollutant emissions. The results indicated that both gaseous pollutants and PM exhibited 21 much higher concentrations during the rush hours in the morning and at night due to 22 vehicle emission. Two peaks of the PM concentration were observed in the scope of 0.7-1.1 23 and 3.3-4.7 µm, accounting for 14.6% and 20.3% of the total concentrations, respectively. 24 Organic matter (OM), EC, and many water-soluble ions were markedly higher at the rush 25 hours in the morning than those at night, implicating comprehensive effects of vehicle 26 types and traffic volume. The particle number concentrations exhibited two peaks at Aitken 27 mode (25 nm and 100 nm) and accumulation mode (600 nm), while the particle volume 28 concentration displayed high values at the accumulation mode (100-500 nm) and coarse 29 mode (2.5–4.0 μm). The peak around 100 nm was detected in the morning rush hours, but it 30 diminished with the decrease of the traffic volume. Individual-particle analysis revealed 31 that main particles in the tunnel were Fe-rich particles, K-rich particles, mineral particles, 32 Ca-S rich particles and Al-Si particles. The particles collected at the rush hours displayed 33 marked different morphologies, element concentrations and particle sizes compared to the 34 ones collected at the non-rush period. The data presented herein could shed a light on the 35 feature of vehicle emissions. 36

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#### 59 Introduction

Aerosol particles play significant roles on climate by changing
the energy transfer through the radiative forcing, and potentially cause adverse health effects as carriers of toxic chemicals
(Li et al., 2008; Lin et al., 2005; Jacobson, 2001). Some aerosol

particles probably absorb or scatter the solar radiation, thereby 55 influencing the global climate (Anenberg et al., 2012; Liu et al., 56 2014). Health studies have proved that fine particulate ( $PM_{2.5}$ ) 57 showed close relationship to respiratory and cardiovascular 58 disease, and even increased risk for cancer or death (Adar et al., 59 2007; Liu et al., 2015). Exposure to organic particles was 60

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associated with allergies and adverse respiratory effects,
frequently expressed as asthma or chronic obstructive pulmonary disease (Lerner et al., 2012).

Vehicle emissions have been considered as a main source of 64 atmospheric pollutants (Chiang and Huang, 2009). About 23% of 65 NO<sub>x</sub>, 17% of CO, and more than 50% of VOCs were released from Q8 67 vehicle emissions in East China (Fu et al., 2013; Chiang et al., 2007; Lee et al., 2002). Moreover, about 6.7 million tons and 68 69 8.3 million tons of PM<sub>2.5</sub> and PM<sub>10</sub> were emitted by vehicles in 70 Pearl River Delta in 2006, respectively (Cui et al., 2016). The 71 occurrence of hazy days has been increasing since 2005, 72 particularly in some megacities of China (Tong et al., 2007). 73 Some gaseous pollutants (e.g., NOx and VOCs) were the key precursors of second species, such as ozone, in ambient air, 74 75 which probably induced a severe fog-haze episode (Zhou et al., 09 2014). S. Guo et al. (2014), L. Guo et al. (2014) concluded that photochemical oxidation of NO<sub>x</sub> and SO<sub>2</sub> emitted from traffic 77 emissions was primarily responsible for urban haze in China. 78 79 Hu et al. (2015) also reported that Zn(NO<sub>3</sub>)<sub>2</sub> and ZnSO<sub>4</sub> displayed higher concentrations in the haze-fog episodes compared with 80 81 the clear episodes. The number fractions of organic carbon 82 principally derived from vehicle emissions reached to 30% in 83 Shanghai (Hu et al., 2016). In recent years, the numbers of 84 automobiles increased from 187 to 296 million between 2011 and 2015 in China, seriously deteriorating air quality (Cai et al., 85 2010). Thus, to address the feature of vehicle exhausts has 86 87 become a crucial issue to understand the influence of vehicle 88 emissions on air quality in China.

89 It is an arduous challenge to investigate the characteristics of 90 vehicle emissions accurately to ambient air because these 91 pollutants emitted from vehicles are inclined to mix with PM rapidly in the atmosphere. To date, many methods have been 92 93 used to address vehicle emission features, including the engine dynamometer test (Na et al., 2015), remote sensing (Andrew, 94 95 et al., 2003), and tunnel research (Liu et al., 2012). Dynamometer test is costly and its procedure is complex. Furthermore, the test 96 condition of the dynamometer cannot absolutely simulate the 97 realistic driving state. Remote sensing just measures emissions 98 of individual vehicle as they drive by a roadside sensor, but not 99 obtain fleet-average emission results. Moreover, the dynamom-100 eter test and remote sensing cannot distinguish the vehicle 101 emissions from other sources effectively. In contrast, tunnel 102 research not only reflects the real driving environment with less 103 environmental interference, but also makes use of the fact that 104 105 emissions from vehicles inside tunnels could be isolated from 106 other sources easily. Additionally, there is an increased emissions in concentration and prolonged exposure to the pollutants 107 108 emitted by the vehicles due to the confined space and inadequate ventilation (El-Fadel and Hashisho, 2001; Brito et al., 2013; Ho 109 et al., 2004). Thus, the research on the tunnel has become a 110 popular mean to study the characteristics of vehicle emissions. 111

A growing body of tunnel studies has been conducted in 112 113 megacities on the world. Many tunnel studies focused on the characteristics of emissions including concentrations, compo-114 115 nents and emission factors of the pollutants. Ma et al. (2011) reported that the concentrations of CO,  $SO_2$ , and  $NO_x$ , reached to 116 12-39 ppm, 20-48 ppb, and 1.2-3.1 ppm, respectively, in the 117 Hsueh-shan tunnel. Ho et al. (2009) reported that the VOC 118 concentrations in Shing Mun tunnel were about 5-10 times 119 higher than those in the ambient air of Hong Kong. It were 120

generally considered that benzene, toluene, xylene and isoprene 121 were the major species of VOCs, all of which generally exhibit 122 higher concentrations in the tunnel due to high intensity 123 emission of vehicle exhausts (Chiang et al., 2007). Carbonaceous 124 compounds and water-soluble ions were primary components 125 of PM collected from the tunnel (Pio et al., 2013). However, the 126 components and concentrations of the pollutants varied signif- 127 icantly in the different tunnels probably due to the impacts of 128 engine type, fuel consumption, vehicle characteristics and other 129 uncertain factors (Alves et al., 2015). It was well documented that 130 the number, speed, age, and type of vehicle showed remarkable 131 effects on the concentrations and species of gaseous pollutants 132 and atmospheric PM inside the tunnel (Shu et al., 2001). Vehicle Q10 number generally showed a significantly positive relevance with 134 the pollutant concentration inside the tunnel (Zhou et al., 2014). 135 Especially, the emission factors in the Wuzushan tunnel were 136 higher than those in the Kuixinglou tunnel as the result of high 137 proportion of diesel vehicle in the former (Cui et al., 2016). The 138 NO<sub>x</sub> emission increased with vehicle speed for uphill driving and 139 possessed higher values for uphill driving than those for 140 downhill driving (Andrew et al., 2003). However, few studies 141 concerned about the morphology and mixing state of PM within 142 the tunnel, which were also sensitive to the vehicle emissions. 143 Knowledge of the morphology and mixing state of individual 144 particles in the tunnel was crucial to understand the vehicle 145 emission features (Li et al., 2016). 146

In the present study, the PM samples were collected and 147 gaseous pollutants were on-line analyzed over a 24-hr period 148 in the Xiangyin tunnel of Shanghai. The main objectives of 149 this study are: (1) to investigate the diurnal variation of 150 gaseous pollutants and PM inside the tunnel, (2) to determine 151 the concentration and composition of PM, and (3) to decipher 152 the number and volume concentrations, size distribution and 153 morphology of PM at a single-particle level. 154

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### 1. Experimental section

The samples were collected in the Xiangyin tunnel, which was 157 located at the middle ring of Shanghai. This tunnel is a length 158 of 2.6 km with the maximum speed limit of 80 km/hr. Diesel 159 and gasoline vehicles were main vehicle types across the 160 Xiangyin tunnel. The proportion of diesel vehicles through 161 Xiangyin tunnel was high in the daytime, while they were 162 relatively low at night because diesel vehicles were prohibited 163 entering downtown area since the evening rush hours. The 164 samples including aerosol particles and gaseous pollutants 165 inside the tunnel were collected at the midpoint of the tunnel, 166 and the ambient samples outside the tunnel were collected in 167 the site about 50 m far from the exit of the tunnel. Ambient air 168 samples in the tunnel were collected once each hour from 169 14:30 on April 24th to 14:30 on April 25th, 2013. The sampling 170 period was a weekday, which could represent the pollution 171 status in the tunnel in the weekdays. Ambient air was 172 collected at a flow rate of 1 L/min. The samples were analyzed 173 within one day after the sampling. 174

Particles inside the tunnel were collected in the eight size 175 fractions: 0.4–0.7, 0,7–1.1, 1.1–2.1, 2.1–3.3, 3.3–4.7, 4.7–5.8, 5.8–9, 176 and >9  $\mu$ m, respectively. The sampling pump flow rate was 177 set at 28.3 L/min. Before the sampling, the quartz filters were 178

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