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Concentrations of ultrafine particles at a highway toll collection booth and exposure implications for toll collectors

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ABSTRACT

Research regarding the magnitude of ultrafine particle levels at highway toll stations is limited. This study measured ambient concentrations of ultrafine particles at a highway toll station from October 30 to November 1 and November 5 to November 6, 2008. A scanning mobility particle sizer was used to measure ultrafine particle concentrations at a ticket/cash tollbooth. Levels of hourly average ultrafine particles at the tollbooth were about 3–6 times higher than those in urban backgrounds, indicating that a considerable amount of ultrafine particles are exhausted from passing vehicles. A bi-modal size distribution pattern with a dominant mode at about <6 nm and a minor mode at about 40 nm was observed at the tollbooth. The high amounts of nanoparticles in this study can be attributed to gas-to-particle reactions in fresh fumes emitted directly from vehicles. The influences of traffic volume, wind speed, and relative humidity on ultrafine particle concentrations were also determined. High ambient concentrations of ultrafine particles existed under low wind speed, low relative humidity, and high traffic volume. Although different factors account for high ambient concentrations of ultrafine particles at the tollbooth, measurements indicate that toll collectors who work close to traffic emission sources have a high exposure risk.

1. Introduction

Defined as particles with diameters <100 nm, ultrafine particles typically form through nucleation, gas-to-particle reactions, or evaporation/condensation. Recent studies have shown increasing interest in concentrations of ultrafine particles at workplaces, as adverse pulmonary health effects are associated with freshly generated ultrafine particles (Oberdörster, 2001; Donaldson et al., 2002; Oberdörster et al., 2002; Englert, 2004; Gilmour et al., 2004). Toxicological and epidemiological studies have also demonstrated that ultrafine particles are more harmful to health than large particles because small particles have a much larger adsorbed or condensed surface area than large particles with the same mass, and can become deposited in alveoli, where they interact with epithelial cells (Jaques and Chong, 2000; Pekkanen et al., 2002; Brown et al., 2002; Delfino et al., 2005; Oberdörster et al., 2005). Ultrafine particles from traffic exhaust can also damage DNA (Bräuner et al., 2007; Møller et al., 2008). Thus, ultrafine particle number concentrations are a threat to occupational/public health.

Occupational exposure to ultrafine particles may be high when workers are close to emission sources. Ultrafine particles in workplaces may originate from several sources such as hot processes, vehicle emissions, combustion processes, and high-speed mechanical processes. For example, ultrafine particles are typically encountered in the workplace when combustion fumes and saturated vapors arise from hot processes such as smelting, welding, soldering, and plasma spraying (Wake et al., 2002). Ultrafine particles can also be generated by such mechanical processes as grinding, cutting and polishing (Zimmer and Maynard, 2002). Many studies have assessed occupational exposure levels of ultrafine particles at different workplaces (Zimmer and Biswas, 2001; Zimmer et al., 2002; Brouwer et al., 2004; Wheatley and Sadhra, 2004; Peters et al., 2006; Thomassen et al., 2006; Heitbrink et al., 2007; Cheng et al., 2008; Demou et al., 2008; Evans et al., 2008; Elihn and Berg, 2009). Wake et al. (2002) noted that ultrafine particle concentrations of $>5.0\times10^5$ particles cm^{-3} were emitted from galvanizing, welding, soldering, and plasma spraying processes. Wheatley and Sadhra (2004) determined that ultrafine particle levels were about 5.8×10^4–2.3×10^5 particles cm^{-3} at nine distribution depots where ultrafine particles were exhausted from diesel-powered forklift trucks. Brouwer et al. (2004) demonstrated that levels of ultrafine particles were $>1.0\times10^5$ particles cm^{-3} from a welding process. Cheng et al. (2008) demonstrated that levels of ultrafine particles were 2.0×10^4–2.8×10^5 particles cm^{-3} at an iron foundry. Evans et al. (2008) found that ultrafine particle levels in an automotive grey-iron foundry were 7.0×10^4–2.8×10^5 particles cm^{-3}. Ultrafine particle levels were about 1.5×10^4–1.3×10^5 particles cm^{-3} in

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seven different industrial plants where fettling, laser cutting, welding, smelting, core fabrication, molding, concreting, grinding, sieving, and washing processes were operated (Elihn and Berg, 2009). These measurement results varied depending on emission sources and processes. Notably, peak ultrafine particle concentrations in workplaces can be several times those outdoor.

Toll collectors working at highway toll stations may be exposed to high levels of ultrafine particles emitted from passing vehicles. Previous studies have demonstrated that exposure levels of PM_{10}, $PM_{2.5}$, and polycyclic aromatic hydrocarbons (PAHs) for toll collectors were 373–454 μg m^{-3}, 653–787 μg m^{-3}, and 8.3–12.3 μg m^{-3}, respectively, at a highway toll station. These high pollutant exposure levels were strongly correlated with traffic volume (Lai et al., 2004; Tsai et al., 2004). However, information is lacking for exposure levels to ultrafine particles for toll collectors at highway toll stations who are directly exposed to ultrafine particles emitted directly from passenger cars. Furthermore, ultrafine particle levels in outdoor environments depend on vehicle emission intensity, meteorological factors, and pre-existing particle concentrations (Charron and Harrison, 2003). Zhu et al. (2002) demonstrated that particle number concentrations declined significantly when wind speed increased downwind of a highway.

Taiwan's highway toll stations currently have two methods for collecting tolls—manual toll collection and electronic toll collection (ETC). However, manual toll collection is still the dominant method at Taiwan's highway toll stations. In this method, a toll collector in a tollbooth collects tickets or cash from passing vehicles.

This study evaluates ambient levels of ultrafine particles at a highway toll collection booth and discusses exposure implications for toll collectors. The influences of traffic volume and meteorological conditions on ultrafine particle concentrations are also investigated via field monitoring.

2. Materials and methods

2.1. Sampling site

The sampling site in this study is a toll station on Highway 1, 10 km west of the Taipei City center (Fig. 1). According to the Bureau of Highway records, this toll station has the highest traffic volume among all toll stations in Taiwan. There are 22 toll collection booths at this station, and the daily traffic volume at this station is about 227,000 vehicles day^{-1}. Eleven of these booths collect tolls from vehicles traveling from Taipei County to Taoyuan County ("north" to "south"), and the remaining eleven booths collect tolls from vehicles traveling in the opposite direction. Eight lanes in each direction are designated for passenger cars (L1–L8), and three lanes are for trucks and buses (L9–L11). Of the eight passenger car lanes, four lanes are for drivers who have tickets (L3–L6), two lanes are for drivers who have tickets or cash (L7–L8), and two lanes are for ETC (L1–L2). The highway toll station runs three work shifts per day—day shift (8:00–16:00), afternoon shift (16:00–24:00), and night shift (00:00–8:00). During working hours, a toll collector stands in a tollbooth with one door open toward the traffic lane and faces the traffic lane when collecting tolls from drivers. This study measured the levels of ultrafine particles at the L7 tollbooth for passenger cars driving north to south because this lane is still open during off-peak hours.

2.2. Sampling equipment and experimental design

Due to a lack of suitable personal samplers for ultrafine particles to assess workplace exposure and risk, studies have typically adopted measurements at fixed locations to determine worker exposure to ultrafine particles. This study used a scanning mobility particle sizer (SMPS) (TSI Model 3936; TSI, Inc., Shoreview, MN, USA) to measure ultrafine particle concentrations. During monitoring, the SMPS was placed on the ticket/cash lane 1 m from the L7 tollbooth. The inlet of the SMPS was located roughly 1.5 m above the ground and facing the traffic lane. In this study, the SMPS measured concentrations of ultrafine particles 6–225 nm in diameter. Each scan was completed by an SMPS up-scan of 120 s, a retrace of 30 s, and a standby of 30 s. Twenty measurements were taken hourly. The SMPS was operated continuously for approximately 23 h during each on-site monitoring period (1:00–24:00). After each monitoring session, the inlet impactor was cleaned and greased. The ultrafine particle number concentrations were measured on five sampling days at the L7 tollbooth from October 30 to November 1 (Thursday to Saturday) and

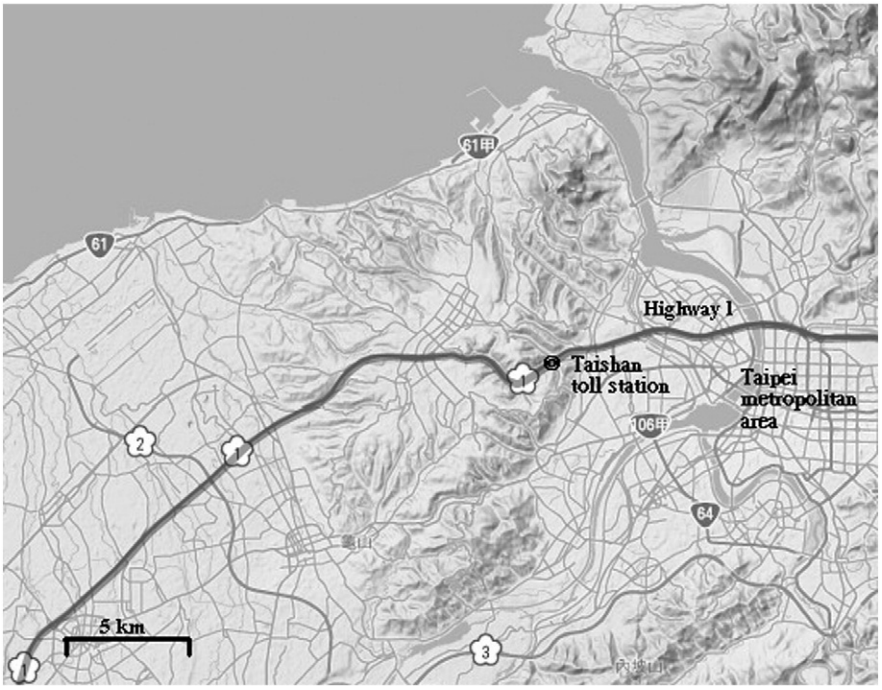

Fig. 1. Schematic diagram of the sampling location.

November 5 to November 6 (Wednesday to Thursday), 2008. The hourly average ultrafine particle number concentrations within the size range of 6–100 nm were calculated from SMPS raw data, which were corrected for diffusion losses inside the SMPS. Hourly traffic volumes were recorded automatically by a sensor and data were obtained from the Toll Station Administration. Local meteorological data were recorded at 10-min. intervals by a Vantage Pro 2™ Weather Station (Davis Instruments, Hayward, CA, USA), set up at the toll plaza.

2.3. Statistical methods

The Pearson product moment correlation coefficient ($R_{Pearson}$) was used to assess the strength of correlations between hourly average ultrafine particle concentrations and hourly traffic volumes. Multivariate regression analyses were conducted using SPSS statistical software to determine hourly average levels of ultrafine particles at the L7 tollbooth based on hourly traffic volumes and hourly average meteorological conditions. The stepwise regression method was applied to identify significant independent variables, such as traffic volume, wind speed, wind direction, temperature, and relative humidity, associated with concentrations of ultrafine particles at the L7 tollbooth. The stepwise procedure allows for the addition of each independent variable into the regression model, from highest to lowest, in order of their individual correlation with the dependent variable. This procedure can circumvent the potential problem of multicollinearity, because a variable will not be entered into the regression model if it is already strongly correlated with a variable in the regression model. An independent sample t-test was also performed to test the magnitude of differences in hourly average ultrafine particle levels, hourly traffic volumes and hourly average wind speeds for different shifts. A significance level of 0.05 was used for all statistical tests.

3. Results and discussion

3.1. Ultrafine particle levels at the tollbooth

Table 1 lists the hourly average ultrafine particle levels at the L7 tollbooth during monitoring periods. The hourly average ultrafine particle levels at the L7 tollbooth were 9.3×10^3–1.6×10^5 particles cm^{-3} (mean = 6.5×10^4 particles cm^{-3}). Under urban backgrounds, the mean number concentrations of ultrafine particles were measured at 1.0×10^4–2.0×10^4 particles cm^{-3} (Li et al., 1993; Tuch et al., 1997; Hughes et al., 1998; Shi et al., 1999; Ruuskanen et al., 2001; Pekkanen et al., 2002; Matson, 2005; Rodríguez et al., 2007). Li et al. (1993) demonstrated that levels of ultrafine particles in urban Taipei were about 7.5×10^3–3.5×10^4 particles cm^{-3} (mean = 1.5×10^4 particles cm^{-3}). Compared with urban background ultrafine particle levels, levels of hourly average ultrafine particles at the L7 tollbooth were about 3–6 times higher, indicating that a considerable amount of ultrafine particles was exhausted directly from idling vehicles. This measurement result is similar to that obtained by Hitchins et al. (2000), who reported that ultrafine particle number concentrations near major roads were up to 3.5–7 times higher than those in urban ambient air. Jayaratne et al. (2009) recently determined that the amount of particles emitted from vehicles increased when vehicles were forced to stop, idle, and accelerate from rest, such as that which occurs at signal intersections and pedestrian crossings. In this study, vehicles in the ticket/cash lane stopped during the toll transaction, idled, and then accelerated when leaving the toll station. Thus, vehicles stopping and then starting from rest and accelerating are potential localized sources of high particle number emissions.

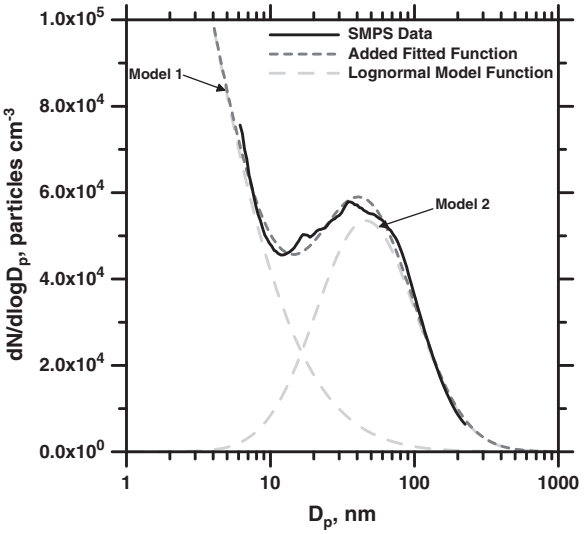

Fig. 2. Average particle number size distribution measured at the L7 tollbooth.

Fig. 2 shows the average particle size distribution measured on five monitoring days at the L7 tollbooth. A size distribution in the range of 6–225 nm was obtained by averaging data from >2200 SMPS scans. A bi-modal size distribution pattern with a dominant mode at <6 nm and a minor mode at about 40 nm was observed at the L7 tollbooth. Based on measurement results, particle number concentrations increased quickly as particle diameters decreased, such as when ultrafine particle diameters were <10 nm. The high amounts of nanoparticles in this study can be attributed to gas-to-particle reactions in fresh fumes emitted directly from vehicles. The measured size distribution was fitted with DistFit software (Chimera Tech., Inc., Forest Lake, MN, USA). The fitting result shows that a bi-modal size distribution pattern can be simulated accurately by two lognormal models. The simulation indicates that this bi-modal size distribution was combined with two individual lognormal size distributions. The count median diameter and geometric standard deviation of the first lognormal size distribution were 0.52 nm and 5.12, respectively. Additionally, the count median diameter and geometric standard deviation of the second lognormal size distribution were 45.95 nm and 2.21, respectively. The ultrafine particles <10 nm in diameter were likely from a gas-to-particle reaction process in fresh fumes emitted from passing vehicles, and ultrafine particles about 30–60 nm in diameter were likely from the condensation process and coagulation growth in ambient air. Measurement results indicate that the mean number concentrations over the measured size range were governed by particles in these two size ranges.

Table 1
Hourly average ultrafine particle levels at the L7 tollbooth.

	Mean[a] (S.D.[b])	Min–max[c]	Median	Q_1–Q_3[d]
UFP[e], particles cm^{-3}	6.5×10^4 (2.7×10^4)	9.3×10^3–1.6×10^5	6.3×10^4	4.8×10^4–7.8×10^4

[a] Observation number N = 115.
[b] S.D.: standard deviation.
[c] Min–max: minimal value–maximal value.
[d] Q_1–Q_3: first quartile value–third quartile value.
[e] Ultrafine particle.

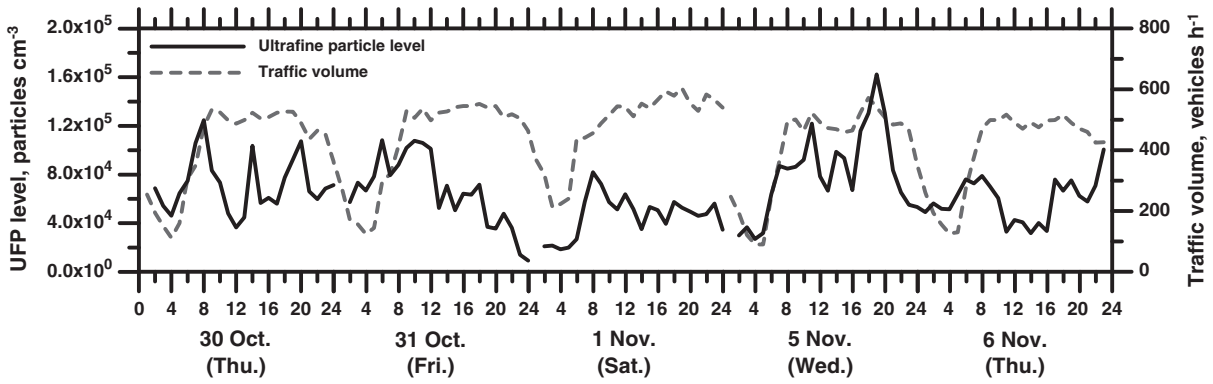

Fig. 3. Time variations of ultrafine particle concentrations and traffic volumes throughout the monitoring period at the L7 tollbooth.

Fig. 3 presents time variations in hourly average ultrafine particle concentrations and hourly traffic volumes during monitoring periods. The ultrafine particle concentrations varied markedly. Furthermore, less variation existed in daytime traffic volumes than in nighttime traffic volumes and low traffic volumes existed during 0:00–5:00 at the L7 tollbooth. Variations in hourly average ultrafine particle levels were positively and weakly correlated with hourly traffic volumes ($R_{Pearson} = 0.20$; $p = 0.017$), indicating that variations in ultrafine particle levels at the L7 tollbooth were caused by traffic emissions and influenced by the local meteorological factors such as wind speed.

3.2. Ultrafine particle levels affected by traffic volumes and meteorological conditions

During monitoring periods, hourly average wind speed was 0.3–2.7 m s^{-1} (mean = 1.1 m s^{-1}); hourly prevalent wind direction was between north and northeast (NNE) and southeast (SE); hourly average temperature was 22.7–33.9 °C (mean = 27.5 °C); and hourly average relative humidity was 50–86% (mean = 72.1%).

Multivariate regression results show that concentrations of ultrafine particles were strongly correlated with traffic volume ($p<0.001$), wind speed ($p<0.001$) and relative humidity ($p = 0.006$), but not with wind direction or ambient temperature, as determined by a stepwise regression method. Table 2 shows the estimated coefficients of the multivariate regression equation for ultrafine particle levels with these three independent variables—traffic volume, wind speed, and relative humidity—at the L7 tollbooth. The R^2 of the multivariate regression equation was 0.48 (adjusted $R^2 = 0.47$). Multivariate regression results demonstrate that levels of ultrafine particles and traffic volume were positively correlated, indicating that levels of ultrafine particles at the L7 tollbooth increased as the number of vehicles increased. That is, traffic is a major contributor of ultrafine particles at the tollbooth. Ultrafine particle concentrations and wind speed were negatively correlated, suggesting that ultrafine particle concentrations at the L7 tollbooth decreased as wind speed increased. That is, atmospheric dilution of ultrafine particles by wind was significant. Similar measurement results for the effect of wind speed on ultrafine particle concentrations were obtained by Charron and Harrison (2003) and Hussein et al.

Table 2
Traffic volume, wind speed, and relative humidity as determinants of ultrafine particle number concentrations by linear regression.

Variable	Estimated coefficient	t-statistic	p value for t-test
Constant	1.31×10^5	4.67	<0.001
Traffic volume, vehicles h^{-1}	99.40	4.90	<0.001
Wind speed, m s^{-1}	-4.09×10^4	−9.51	<0.001
RH, %	-8.59×10^2	−2.80	0.006
F-statistic	34.19		
p value for F-test	<0.001		
R^2	0.48		
Adjusted R^2	0.47		

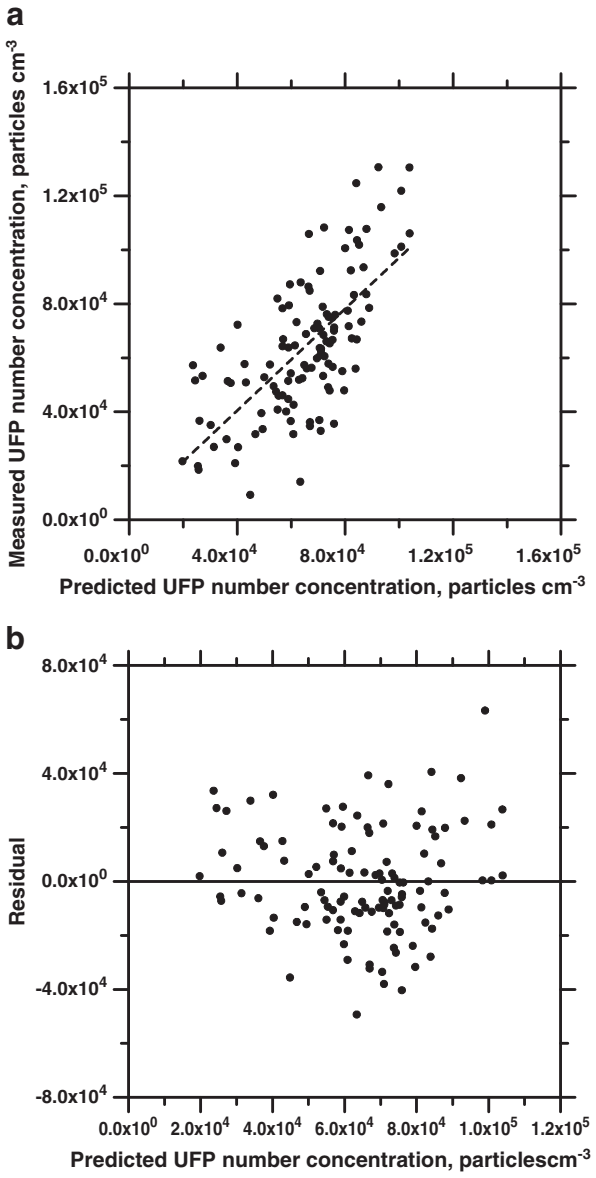

Fig. 4. Relationships between predicted ultrafine particle levels and measured ultrafine particle levels during the entire sampling period (a), and relationships between predicted ultrafine particle levels and residuals (b).

(2005). Moreover, ultrafine particle concentrations and relative humidity were negatively correlated, suggesting that ultrafine particle concentrations decreased, as ultrafine particles easily become large particles via condensation or coagulation under high relative humidity. Additionally, wind direction did not significantly affect ultrafine particle concentrations, because ultrafine particle concentrations in this study were measured close to their emission source (<1 m) at the L7 tollbooth, unlike previous researchers who measured ultrafine particle concentrations at the roadside or at a downwind site. This accounts for why nanoparticles in fresh fumes were observed in this study. Moreover, Hussein et al. (2005) noted that temperature (250–300 K) has a significant effect on ultrafine particle levels. However, ultrafine particle levels were not strongly correlated with ambient temperature (23–34 °C) in this study.

Fig. 4a and b shows the relationships between predicted ultrafine particle levels and measured ultrafine particle levels throughout the entire measurement period, and the relationships between predicted ultrafine particle levels and residuals. Measurement data were in good agreement with values predicted by the multivariate regression equation with traffic volume, wind speed, and relative humidity at 90–600 vehicles h^{-1}, 0.3–2.7 m s^{-1}, and 50–86%, respectively. The residual plot shows that residual distribution is approximately straight and flat, suggesting this multivariate regression model is appropriate. According to measurement results, high levels of ultrafine particles existed at low wind speeds, low relative humidity and high traffic volumes.

3.3. Ambient levels of ultrafine particles during different shifts

Fig. 5a–d shows ultrafine particle levels, traffic volumes, wind speeds, and relative humidity for different shifts during the entire measurement period. Traffic volumes were not significantly different between the day shift and afternoon shift ($p = 0.600$); however, traffic volume during the night shift was significantly lower than that during the day shift and afternoon shift ($p < 0.001$ for day shift vs. night shift and afternoon shift vs. night shift). However, measurement results demonstrate that hourly average ultrafine particle levels during the day shift, afternoon shift, and night shift were 6.6×10^4 particles cm^{-3}, 6.7×10^4 particles cm^{-3}, and 6.1×10^4 particles cm^{-3}, respectively. Statistical results show that ultrafine particle concentrations during these three shifts were not significantly different during the sampling period ($p = 0.953$ for day shift vs. afternoon shift; $p = 0.367$ for day shift vs. night shift; $p = 0.401$ for afternoon shift vs. night shift). Analytical results demonstrate that

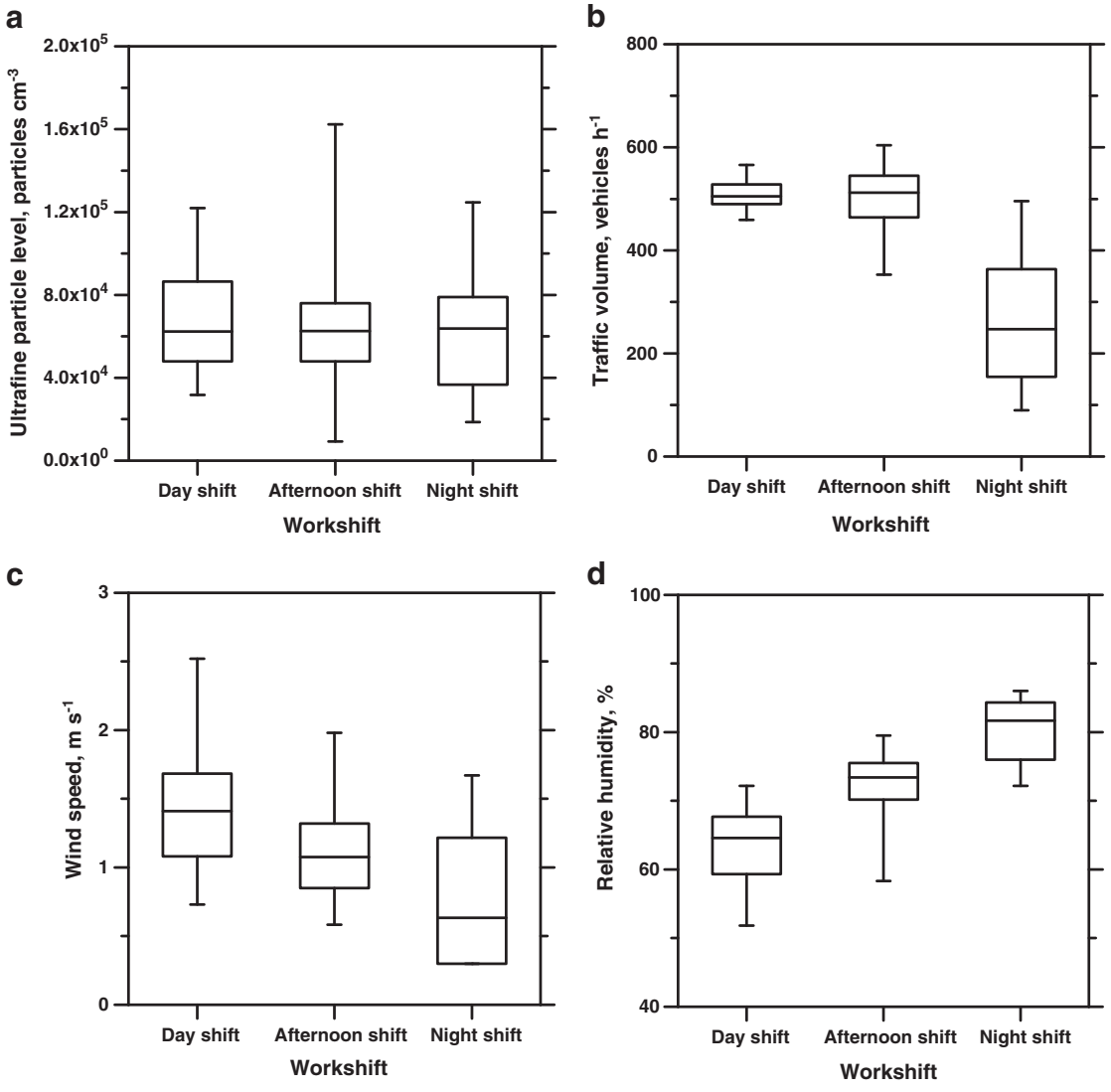

Fig. 5. Ultrafine particle concentrations, traffic volumes, wind speeds, and relative humidity for different shifts throughout the entire measurement period. The box and whisker plot show the lowest, lower quartile, median, upper quartile, and maximum values.

ultrafine particle concentrations at the L7 tollbooth were associated with traffic volume and influenced by local meteorological conditions. High ultrafine particle concentrations still observed under low traffic volumes during the night shift, likely due to the low wind speed. Ultrafine particle levels increased as wind speed decreased, because of reduced ventilation. Moreover, relative humidity was high during the night shift. However, the effect of relative humidity on ultrafine particle concentrations compared to that of wind speed was small, suggesting that ultrafine particle concentrations were dominated by low wind speed during the night shift.

Briggs et al. (2008) noted that the average measured ultrafine particle concentrations while walking in London City was 3.0×10^4 particles cm^{-3}, compared with 2.2×10^4 particles cm^{-3} in a car, suggesting that ultrafine particle exposure while walking on road was substantially higher than that in a car. Compared with measurements acquired by Briggs et al. (2008) for ultrafine particle exposure levels while walking, the ambient level of ultrafine particles for toll collectors was about 2 times higher than those while walking on road. Lai et al. (2004) reported that $PM_{2.5}$ exposure levels for toll station workers were about 110 μg m^{-3}. However, Cheng and Li (2010) reported that ambient levels of $PM_{2.5}$ at same toll station were about 60 μg m^{-3}. These measurment results indicate the $PM_{2.5}$ concentrations inside a tollbooth are about 2 times those around the tollbooth. That is, ultrafine particle concentrations in the tollbooth can be higher than those in ambient air around a tollbooth due to poor tollbooth ventilation. These comparisons indicate that exposure risk is high for highway toll collectors who are directly exposed to traffic emission sources.

4. Conclusions

Ambient concentrations of ultrafine particles for toll collectors were in the range of 9.3×10^3–1.6×10^5 particles cm^{-3} (mean = 6.5×10^4 particles cm^{-3}). This finding demonstrates the effects of traffic volume, wind speed, and relative humidity on ultrafine particle concentrations. High ambient concentrations of ultrafine particles were observed under low wind speed, low relative humidity, and high traffic volume. Ambient levels of ultrafine particles for night shift workers were not significantly different from those for day shift workers and afternoon shift workers due to the low wind speeds during the night shift, even though traffic volumes during the night shift were significantly lower than those during the day shift and afternoon shift. Despite different factors account for high ambient concentrations of ultrafine particles at the L7 tollbooth, toll station workers exposed to high ambient levels of ultrafine particles are at an acute health risk. To reduce health risk, the ETC service has been promoted since 2006 by the Taiwan Area National Freeway Bureau. Oil consumption will be reduced and air and noise quality improved when passenger cars do not stop to pay a toll.

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