Aerosol and Air Quality Research

Source Quantification of PM_{10} and $PM_{2.5}$ Using Iron Tracer Mass Balance in a Seoul Subway Station, South Korea

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ABSTRACT

In this study, we simultaneously measured the PM_{10} and $PM_{2.5}$ mass concentrations and their heavy metal content for three days at a subway station in Seoul to investigate the airborne PM flows. The average concentrations were 59 μg m^{-3}, 37 μg m^{-3}, 111 μg m^{-3}, and 369 μg m^{-3} for the PM_{10} and 43 μg m^{-3}, 28 μg m^{-3}, 58 μg m^{-3}, and 132 μg m^{-3} for the $PM_{2.5}$ at the outdoor air inlet, in the concourse, on the platform, and in the tunnel, respectively. We also found strong correlations between the temporal variations at adjacent sampling locations for both fractions, although they were higher for the $PM_{2.5}$. Additionally, of the airborne trace metals detected at the sampling locations inside the station (the concourse, platform, and tunnel), iron (Fe) displayed the highest concentration and was thus selected as a tracer of PM. Applying a simple mass balance model to the Fe concentrations and ventilation rates revealed that 78% of the PM_{10} and 62% of the $PM_{2.5}$ on the platform emanated from the tunnel, whereas 84% of the PM_{10} and 87% of the $PM_{2.5}$ in the concourse originated outdoors (and arrived in the filtered air). These results further confirm that reducing PM emission from the tunnel is the most effective strategy for improving air quality on the platform and achieving compliance with the national guideline.

Keywords: Subway, Mass balance model, Air quality, Particle matter, Heavy metal

1 INTRODUCTION

The subway is a major public transportation in most megacities throughout the world. For convenience, the subway is usually located in high-density traffic areas with large numbers of pedestrians. People in subway are more prone to be exposed harmful levels of air pollutants if indoor air quality in subway system is not properly managed. Shen and Gao (2019) investigated a personal exposure to PM during four transportations (subway, bicycle, bus and walking) commuting in Nanjing and found that passengers in subway station are exposed to highest $PM_{2.5}$.

The air quality of subway station is largely dependent to characteristics of location and space. Figueroa-Lara *et al.* (2019) found that higher concentration of PM in deeper subway station. PM levels inside the trains and platform decreased with the passage in aboveground sections (Cheng *et al.*, 2011; Carteni *et al.*, 2015; Martins *et al.*, 2016a). Subway air quality is also highly related to the operation conditions (Moreno *et al.*, 2014). The PM concentration increases on a subway environment with an increase of train frequencies (Raut *et al.*, 2009; Colombi *et al.*, 2013; Kwon *et al.*, 2015; Pan *et al.*, 2019) and Woo *et al.* (2018) proposed a model that predicts PM in a subway tunnel as function of train operation.

Since subway PM is mainly generated by friction of rail and cables during train operation,
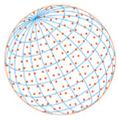
heavy metal content in subway PM is usually high. Park *et al.* (2012) identified that railroad-related sources contributed the most PM_{10} in subway cabin (47.6%) and iron (Fe), manganese (Mn), chromium (Cr) and copper (Cu) are indicators of railroad-related PM_{10} sources. Among all metals, Fe is the most abundant in subway PM (Murruni *et al.*, 2009; Mugica-Avarez *et al.*, 2012; Querol *et al.*, 2012; Loxham *et al.*, 2013; Park *et al.*, 2014; Martins *et al.*, 2016b; Chen *et al.*, 2017; Moreno *et al.*, 2018; Figueroa-Lara *et al.*, 2019).

Moreno *et al.* (2015) found that subway particles are coarser than in buses, trams or outdoor. Qioa *et al.* (2015) found the ratios of PM_1/PM_{10} and $PM_{2.5}/PM_{10}$ in subway are low when the subway train is operating. Son *et al.* (2013) found that particle size of tunnel PM ranged from 1.8 to 5.6 μm. However, smaller size of particles also generated by friction (Midander *et al.*, 2012). Lee *et al.* (2018) found that the size distribution of wear particles generated under the cabin during deceleration was estimated to be bimodal at 165.5 nm and 6.98 nm.

Subway air quality is affected by the outdoor air quality as well as internal sources. Pan *et al.* (2019) found strong linear correlation ($R^2 = 0.897$) between PM in subway station and outdoor PM. The screen doors in the platform block the tunnel PM entering the platform to some extent. It was confirmed that PM concentration on platform effectively decreased after installing platform screen door (Jung *et al.*, 2012; Kim *et al.*, 2012; Yim *et al.*, 2014). Other studies also show that $PM_{2.5}$ concentrations were effectively reduced both in old and new subway stations (Kam *et al.*, 2011; Martin *et al.*, 2015; Minguillon *et al.*, 2018).

The purpose of this study is to quantify the PM sources in a subway station in Seoul by determining PM and air mass balances. Furthermore, this study focused on identification of effective reduction strategy for PM to improve air quality in subway stations.

2 METHODS

2.1 Investigated Station and Sampling Points

Sampling was conducted at a subway station of Seoul Metro Line 4, which is located in the north of Seoul and used by 77 thousand people a day (Fig. 1(a)). We measured PM and heavy metal concentrations between the 14th and 16th of November 2018, taking 13 h air samples between 07:00 to 20:00 for each day. However, measurement could not conduct due to malfunction of sampling equipment at 14 November 07:00–09:00 and 16 November 11:00–14:00.

As shown in Fig. 1(b), we collected samples in four major sectors of the station: outdoor, concourse, platform, and tunnel. The outdoor sampling point was located at the outdoor air inlet
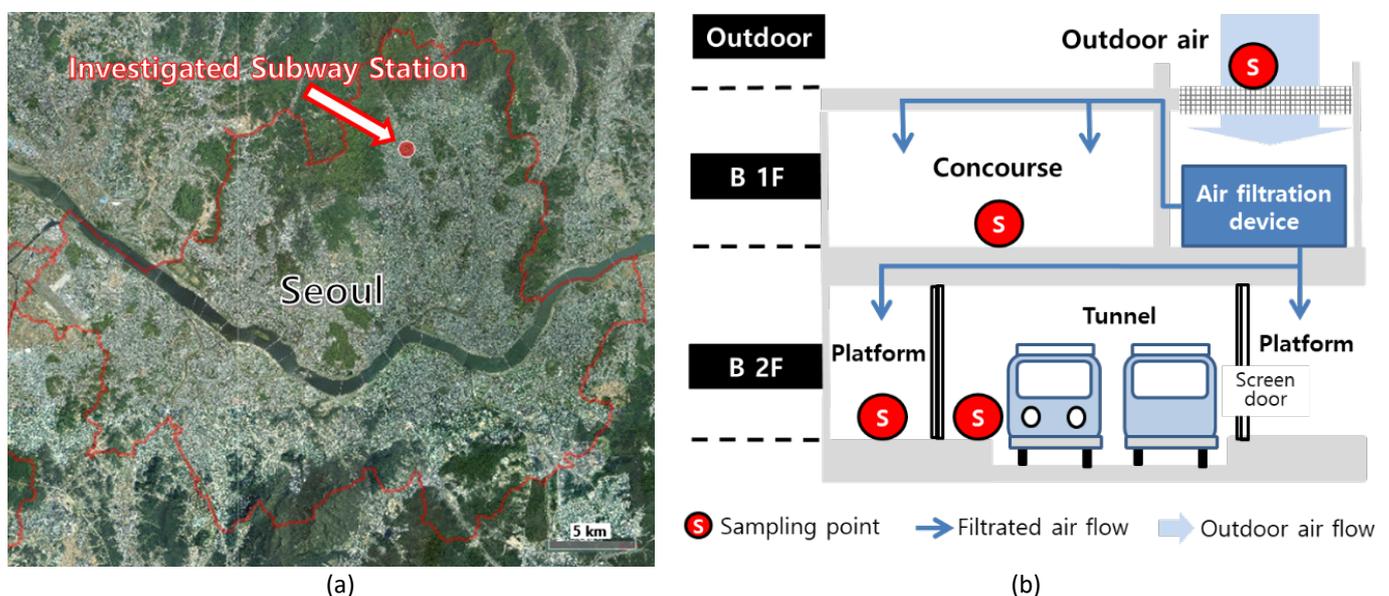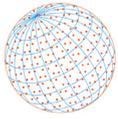

Fig. 1. (a) Location of the subway station within Seoul, South Korea, and (b) schematic of the subway station floor plan and sampling points.

of the air filtration system to ensure the same characteristics of the inflow air into the subway station. In concourse and platform, measurements were taken in the middle sections and at 1.5 m from the ground. Tunnel PM inflowing to the platform was sampled at the rear end of the platform.

$PM_{2.5}$ and PM_{10} were sampled at 6 L min^{-1} and 5 L min^{-1}, respectively, using a portable PM sampler (MiniVol Air Sampler; Airmetrics, USA). The sampling filter was weighed by the auto-weighing system (Chabal-500; C2K Creative, Korea) after filter conditioning for 24 h (temperature 20 $\pm$ 2°C, relative humidity 35 $\pm$ 5%).

As shown in Fig. 1(b), filtrated outdoor air was supplied with rate of 2,384 m^3 min^{-1} in the platform and 1,634 m^3 min^{-1} in concourse. This air filtration system was operated continuously during train operation hours (05:30–00:40). The indoor volume of the platform and concourse was 7,193 m^3 and 11,853 m^3, respectively. The filtrated air was designed to refresh the concourse with 20 times h^{-1} and platform with 8 times h^{-1}. The air filtration device efficiency of PM_{10} and $PM_{2.5}$ were found to be 37% and 35% considering to reduction of PM concentration in filtrated airs. These efficiencies were consistent with other studies for PM_{10} of 30–60%, and $PM_{2.5}$ of 20–40% (Park *et al.*, 2013).

2.2 PM Concentration and Metal Content Analysis

We measured the real-time PM concentration using a light scattering analyzer (Model 1.180; GRIMM, Germany) along with gravimetric method sampling. As the gravimetric method cannot resolve short-term real-time PM variation, the light scattering method was to measure the hourly PM concentrations. Because subway PM has a relatively high Fe content, the light scattering measurements were corrected by the gravimetric method in daily basis.

As shown in Fig. 2, $PM_{2.5}$ and PM_{10} sampled indoor and outdoor in the subway station were extracted using microwaves (QWave 2000; Questron Technologies Corp, Canada) with a 10 mL acid solution (16.7% HCl + 2.5% HNO$_3$). The extract was filtered using a Teflon syringe filter (0.45 μm) and mass up to 25 mL. The pretreatment solution was analyzed using inductively coupled plasma optical emission spectrometry (ICP-OES; CIROS VISION; SPECTRO, Germany) to determine the heavy metal content in the PM. Target heavy metals (Fe, Cu, Cr, Mn) were indicators of railroad-related sources found in Park *et al.* (2012).

3 RESULTS AND DISCUSSION

3.1 PM Concentrations in Subway Station

Fig. 3 shows mean PM_{10} and $PM_{2.5}$ mass concentrations outside and inside subway station measured during the study period. The average PM_{10} concentration were 59 μg m^{-3}, 37 μg m^{-3}, 111 μg m^{-3} and 369 μg m^{-3} in outdoor, concourse, platform and tunnel. And the average $PM_{2.5}$ concentrations were 43 μg m^{-3}, 28 μg m^{-3}, 58 μg m^{-3} and 132 μg m^{-3} in outdoor, concourse, platform and tunnel. In concourse, concentration of PM_{10} and $PM_{2.5}$ were 37% and 35% lower than in outdoor. However, in platform, the PM_{10} and $PM_{2.5}$ concentrations were higher by 88% and 35% than the outdoor, despite with larger the filtered air supply. This occurred because of the significant PM entrainment from the tunnel even with screen installed. The concentrations of PM_{10} and $PM_{2.5}$ in tunnel were more than 3 times higher than those in platform, which indicated that considerable portions of PM prevented from entering through platform from tunnel loaded with heavy PM.

The ratios of $PM_{2.5}/PM_{10}$ on measurement sites in subway station are shown in Table 1. The ratio in tunnel was lowest and consistent with previous studies (Son *et al.*, 2013; Cusack *et al.*, 2015; Qioa *et al.*, 2015). In concourse, level of $PM_{2.5}/PM_{10}$ concentration ratio was similar to those of outdoor due to low influence from tunnel PM. As the platform air was affected by both outdoor and tunnel, the $PM_{2.5}/PM_{10}$ concentration ratio in platform was placed half-value between them.

3.1 Temporal Correlation of PM among Subway Sampling Locations

Table 2 shows the temporal correlations of PM_{10} and $PM_{2.5}$ among sampling sites, namely outdoor, concourse, platform and tunnel. All correlation coefficients of PM between sampling locations were very large (r > 0.8). Particularly, correlations between adjacent locations were
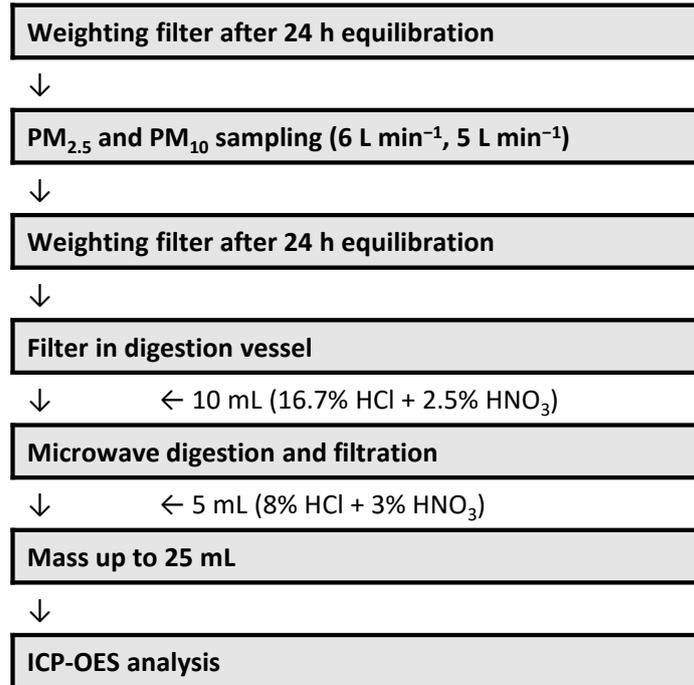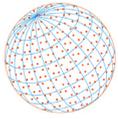
Weighting filter after 24 h equilibration

↓

PM$_{2.5}$ and PM$_{10}$ sampling (6 L min^{-1}, 5 L min^{-1})

↓

Weighting filter after 24 h equilibration

↓

Filter in digestion vessel

↓ ← 10 mL (16.7% HCl + 2.5% HNO$_3$)

Microwave digestion and filtration

↓ ← 5 mL (8% HCl + 3% HNO$_3$)

Mass up to 25 mL

↓

ICP-OES analysis

Fig. 2. Pretreatment and analytical procedure for the determination of heavy metal composition.
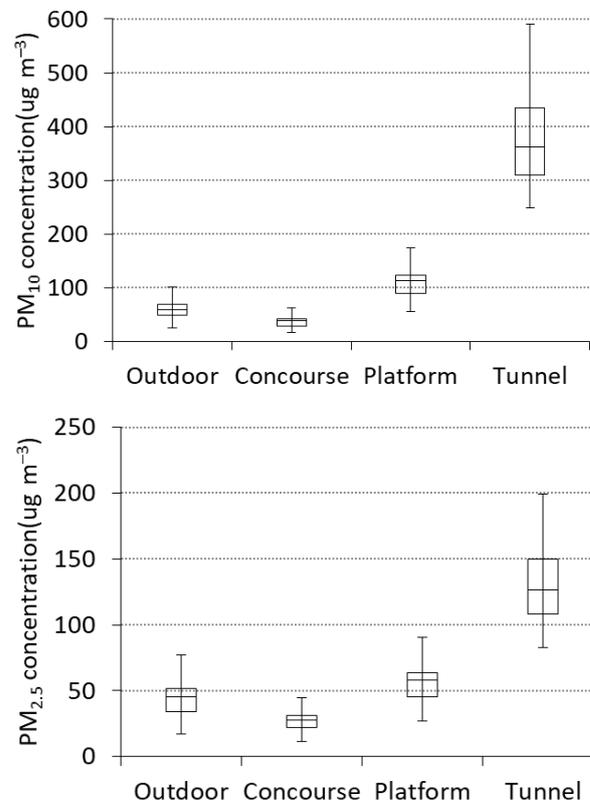

Fig. 3. PM concentration outside and inside subway station.

Table 1. Ratio of PM$_{2.5}$ and PM$_{10}$ concentration according to measurement point.

	Outdoor	Concourse	Platform	Tunnel
PM$_{2.5}$/PM$_{10}$ concentration ratio	0.72 ± 0.03	0.75 ± 0.03	0.52 ± 0.02	0.36 ± 0.02
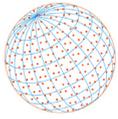
Table 2. PM concentration relationship between the measurement points.

	PM_{10}				$PM_{2.5}$			
	Outdoor	Concourse	Platform	Tunnel	Outdoor	Concourse	Platform	Tunnel
Outdoor	1.00				1.00			
Concourse	0.98	1.00			0.99	1.00		
Platform	0.95	0.98	1.00		0.97	0.98	1.00	
Tunnel	0.82	0.87	0.86	1.00	0.89	0.91	0.91	1.00

close to 1 with exception between platform and tunnel, which indicated active air and aerosol exchanges between different subway locations. Noticeably, the correlations of $PM_{2.5}$ between different sampling locations was slightly higher than that of PM_{10}, which may indicate $PM_{2.5}$ penetration rate was likely higher during the air transfer because of its smaller deposition rate. Therefore, $PM_{2.5}$ would be more suitable to trace its mass budget than PM_{10} with safely ignoring its sinks.

3.2 Heavy Metal Content in PM

Table 3 shows the heavy metal content rate in PM, Fe is the most abundant which were 48% of PM_{10} and 44% of $PM_{2.5}$ in the tunnel. The Fe content of PM_{10} was higher than that of $PM_{2.5}$, which was consistent with the characteristics of relatively higher coarse PM concentration in tunnel. The Fe and Cu contents of PM decreased rapidly in the platform, concourse, and outdoor sampling locations as they were generated in the tunnel, inflowing to the platform and then to the concourse. The ratios of Cu and Fe contents in PM were approximately $\sim$0.03 in regardless of the measurement points, which confirmed that they were originated from common source. According to Park (2013), the annual metal wear amount of Fe and Cu for the Seoul subway lines 1–4 (311 km section) are approximately 17.7 and 0.9 tons (Cu/Fe $\approx$ 0.05) based on the replacement quantity of consumables relevant to train friction. We applied Fe concentration along with PM_{10} and $PM_{2.5}$ mass to estimation of PM contributions from tunnel source to platform and concourse in the next section.

3.3 Mass Balance of PM and Air Flow in the Subway Station

In order to quantify the air flow rates and fluxes of PM (PM_{10}, $PM_{2.5}$) among subway major sectors (tunnel, platform, concourse, and outdoor), a simple mass balance model was implemented as depicted in Fig. 4. This model could fully resolve the air flow rates between subway sectors and how PM and Fe have been distributed among sectors with known information (observed concentrations, ventilation rate of outside airs). The air flow rates in the subway station are expressed as Q (m^3 min^{-1}), where Q_{pc} (air flow rate from platform to concourse), Q_{tp} (tunnel to platform), Q_{c_out} (exhausted air to outdoor from the concourse) and Q_{p_out} (exhausted air to outdoor from the platform) are unknown and to be determined, while Q_{foc} and Q_{fop} are known as they were set by ventilation system with filtration device. The mean concentrations at each sampling point are expressed as C (μg m^{-3}), where C_o, C_c, C_p, C_t can be the concentrations for PM_{10}, $PM_{2.5}$ and Fe in outdoor, concourse, platform, and tunnel.

Table 3. Heavy metal composition of PM in S subway station.

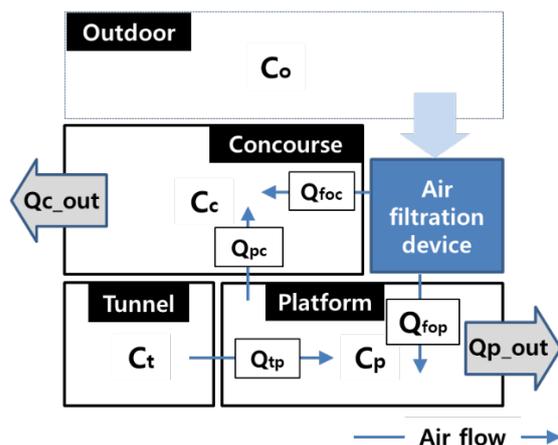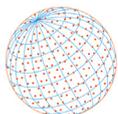| Pollutant	Sampling point	Heavy metal content (%)			
		Fe	Cu	Cr	Mn
PM_{10}	Outdoor	4.6	0.1	0.1	0.1
	Concourse	13.8	0.4	0.2	0.2
	Platform	37.6	1.1	0.1	0.3
	Tunnel	48.4	1.6	0.1	0.4
$PM_{2.5}$	Outdoor	4.0	0.1	0.1	0.1
	Concourse	6.3	0.2	0.1	0.1
	Platform	24.5	0.7	0.1	0.3
	Tunnel	43.5	1.1	0.1	0.3

Fig. 4. Schematic of air flow in the subway station.

To simply this model in estimating four unknown air flow rates (Q_{pc}, Q_{tp}, Q_{c_out}, Q_{p_out}), the following assumptions were made. First, aerosol from outdoor and railway tunnel are only sources of PM and Fe. This assumption is particularly true for Fe, and therefore Fe was used specifically as a tracer for this mass balance models. Second, air flow rates between sectors and aerosol concentrations are steady state throughout the observation period. The aerosol and Fe concentrations of each sector was safely assumed to be a steady state because the residence times of air in concourse and platform are short enough (less than 7 minutes) to achieve steady state.

Aerosol mass balance and air flow balance within concourse were determined as Eqs. (1) and (2), respectively. Also, corresponding balances within platform were determined as Eqs. (3) and (4). η in Eqs. (1) and (3) represents the known collection efficiency of PM for air filtration device in subway ventilation system. The mean Fe concentrations were used as mass tracer in PM$_{2.5}$ in the outdoor (C_o), concourse (C_c), platform (C_p), and tunnel (C_t) which were 1.8 µg m^{-3}, 2.0 µg m^{-3}, 15.3 µg m^{-3} and 55.8 µg m^{-3}, respectively.

$$C_o \cdot (1 - \eta) \cdot Q_{foc} + C_p \cdot Q_{pc} = C_c \cdot Q_{c\_out} \tag{1}$$

$$Q_{foc} + Q_{pc} = Q_{c\_out} \tag{2}$$

$$C_o \cdot (1 - \eta) \cdot Q_{fop} + C_t \cdot Q_{tp} = C_p \cdot Q_{p\_out} \tag{3}$$

$$Q_{fop} + Q_{tp} = Q_{p\_out} \tag{4}$$

Employing Fe concentrations in PM$_{2.5}$ and known η, Q_{foc} and Q_{fop}, four unknown flow rates (Q_{pc}, Q_{tp}, Q_{c_out} and Q_{p_out}) were calculated and listed in Table 4. The result shows that the 94% (1,634 m^3 min^{-1}) of concourse air was originated from ventilation system with outdoor air filtration device and the rest 6% (102 m^3 min^{-1}) was originated from platform air. Although the air flow into concourse from platform was very limited, aerosol mass contribution to concourse from platform were relatively high with 16% (0.7 of total 4.3 g h^{-1}) for PM$_{10}$ and 13% (0.4 of total 3.1 g h^{-1}) for PM$_{2.5}$ due to their higher concentrations in platforms than outdoor air. In case of platform, contributions of air flow rate from ventilation system was lower (74%) than those in concourse although its flow rate of 2,384 m^3 min^{-1} was 46% higher. Increased air flow rate from tunnel with very high PM concentrations, aerosol mass contribution rates from tunnel to platform was very high for 78% (18.4 of total 23.7 g h^{-1}) for PM$_{10}$ and 62% (6.6 of total 10.6 g h^{-1}) for PM$_{2.5}$.

3.4 Verification of Mass Balance Model

To verify that our Fe tracer utilized mass balance model is adequate to reproduce of PM$_{10}$ and PM$_{2.5}$ mass behaviors in each subway sector, PM$_{10}$ and PM$_{2.5}$ mass variations were calculated in concourse and platform using calculated air flow rates, measured PM$_{10}$ and PM$_{2.5}$ in outdoor and tunnel airs during the observation period. The measured and calculated concentrations of PM$_{10}$ and
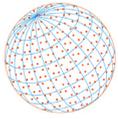
PM$_{2.5}$ in concourse and platforms were compared in Fig. 5. As Fig. 5 shows that calculated PM for each sector successfully reproduced observed values throughout the entire period. The calculated mean concentrations of PM$_{10}$ and PM$_{2.5}$ in the concourse were 41 μg m^{-3} and 29 μg m^{-3}, while the actual concentrations were 37 μg m^{-3} and 28 μg m^{-3}, respectively. The mean differences between the calculated and measured concentrations were 11% and 4% for PM$_{10}$ and PM$_{2.5}$, respectively. The calculated mean concentrations of PM$_{10}$ and PM$_{2.5}$ in the platform were 123 μg m^{-3} and 55 μg m^{-3} respectively, while the measured were 111 μg m^{-3} and 58 μg m^{-3}. 11% and 5% difference for PM$_{10}$ and PM$_{2.5}$ between the calculated and measured concentrations in the platform were very

Table 4. Estimated air and PM flow rates between the subway station sectors.

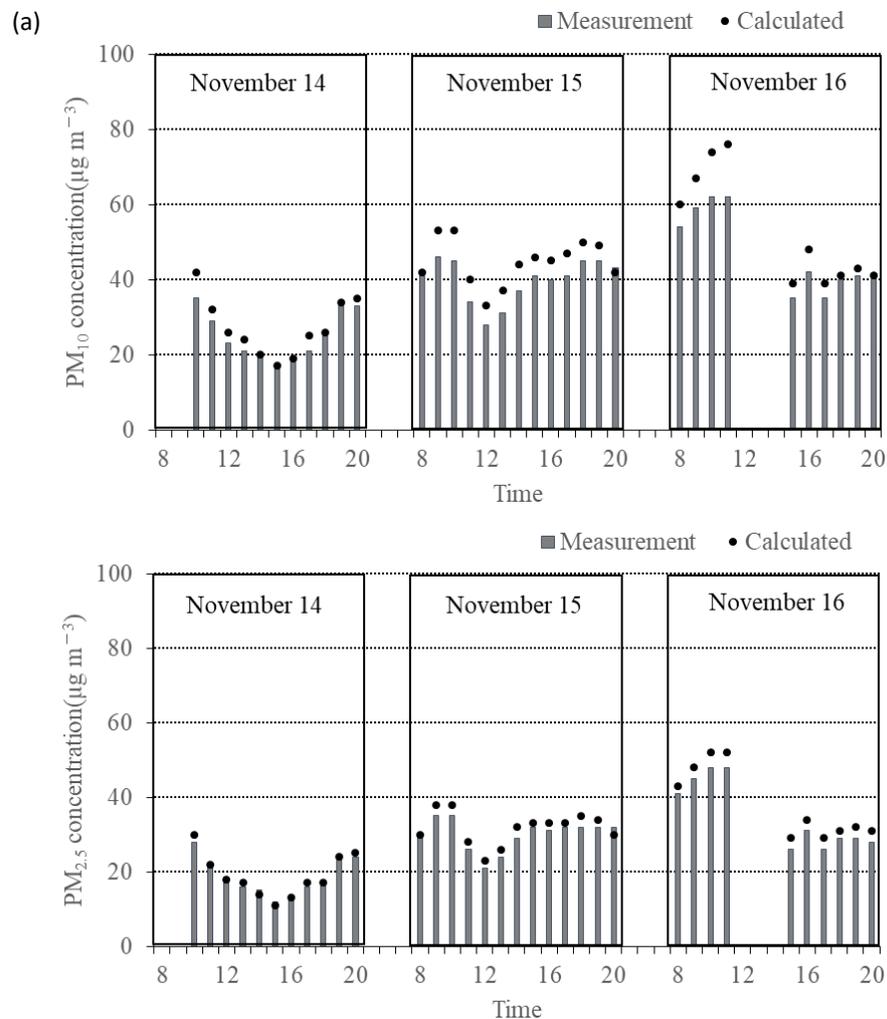| | Flow direction | Air flow rate (m^3 min^{-1}) | PM mass rate (g h^{-1}) | |
			PM$_{10}$	PM$_{2.5}$
Q$_{pc}$	Platform → Concourse	102	0.7 ± 0.2	0.4 ± 0.1
Q$_{foc}$*	Air filtration device → Concourse	1,634	3.6 ± 1.3	2.7 ± 0.9
Q$_{c\_out}$	Concourse → Outdoor	1,736	3.9 ± 1.2	2.9 ± 0.9
Q$_{tp}$	Tunnel → Platform	832	18.4 ± 4.2	6.6 ± 1.5
Q$_{fop}$*	Air filtration device → Platform	2,384	5.3 ± 2.0	4.0 ± 1.4
Q$_{p\_out}$	Platform → Outdoor	3,216	21.4 ± 6.4	11.2 ± 3.5

* known ventilation air flow rate.

Fig. 5. Measured PM concentrations and calculated concentrations in the subway station: (a) concourse and (b) platform.
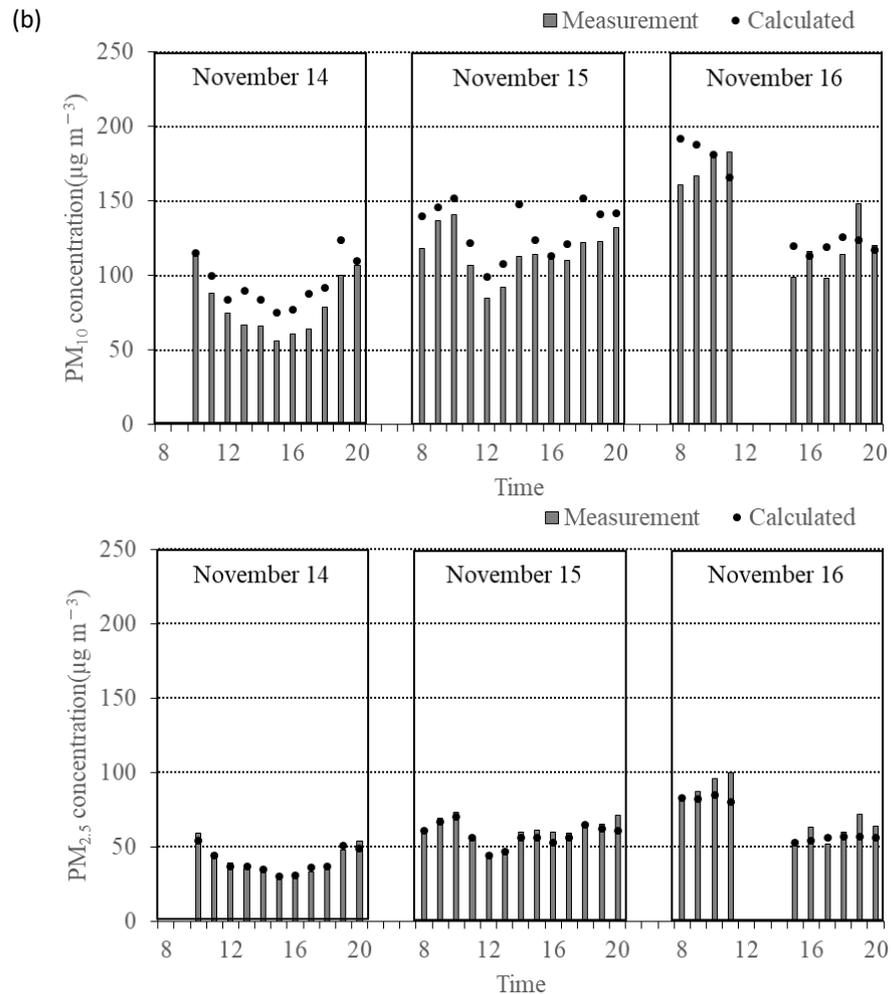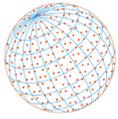
(b)

Fig. 5. (*continued*).

similar to those in the concourse, which indicated that our model and outcomes are sufficiently accurate to quantify behaviors of aerosol transfer between subway sectors.

3.5 Effective Reduction of Subway PM Concentrations

Using the mass balance equations earlier stated, we could assess how to effectively reduce the PM levels in the subway station. The simplest way to improve indoor air quality of subway is to supply cleaner air flows to underground spaces. Fig. 6(a) shows the projecting reduction of PM concentration in the concourse and platform according to PM filtration efficiency of ventilation system. Especially, the PM reduction in concourse was steeply linear to PM filtration efficiency by ventilation system. If the filtration efficiency is improved from current 35% to 70%, the PM_{10} and $PM_{2.5}$ levels in concourse are reduced by 45–50%, which is quite significant. However, the PM_{10} and $PM_{2.5}$ in platform is to reduce only 12% and 20%, respectively along with the same amount of filtration efficiency improvement. The obvious reason was that air in platform was more influenced by highly loaded aerosols from tunnel airs. If the tunnel PM concentration is reduced by half, platform PM_{10} and $PM_{2.5}$ concentrations are reduced to approximately 39% and 31%, respectively (Fig. 6(b)). Consequently, the effective way to reduce PM levels in platform is to regulate subway tunnel aerosol sources. As full-height enclosed screen doors are installed in the platform of studied subway station, platform air is only intermittently exposed from tunnel airs when the trains stop and screen doors are open. Further airtight enhancement between the platform and the train, and tunnel and/or reduction of PM generations from train operations are most direct and effective way to reduce high PM levels in platform. Nevertheless, in concourse, even if the tunnel PM was completely removed, the PM_{10} and $PM_{2.5}$ concentrations are reduced
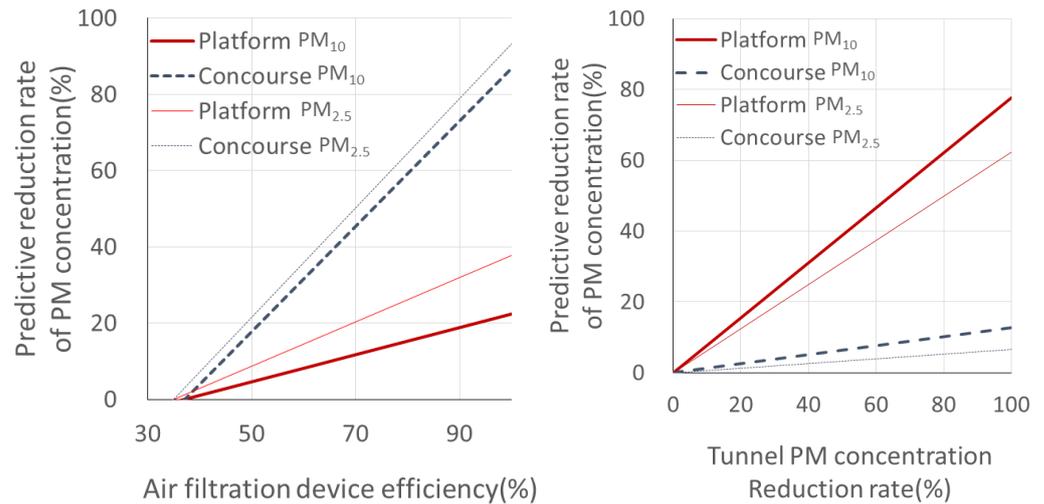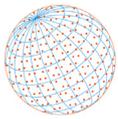
Fig. 6. Predictive reduction rate of PM concentration according to tunnel PM concentration and air filtration device efficiency.

by only 13% and 7%, respectively. It was clearly stated that reduction strategies of PM concentration should be set based on the target station sector.

During the study period, indoor PM levels (average of concourse and platform) were kept below Korean indoor air quality guideline (24 h mean concentration PM_{10}: 100 μg m^{-3}, $PM_{2.5}$: 50 μg m^{-3}). However, hourly $PM_{2.5}$ frequently exceeded 24 h guideline level with peak of 100 μg m^{-3} in platform. To keep the highest peak of $PM_{2.5}$ in the platform under the national guideline, this mass balance approach implied that up to 80% reduction of PM from tunnel source is required if all other conditions remain constant. In order to improve PM levels in the platform, the controlling PM inputs from tunnel should be set in priority as other scheme, such as extra ventilation is not so effective to improve air quality in the platform. This mass balance approach was useful to assess the air quality improvement in the subway spaces and would be adequate to apply the specific reduction strategy plan for subway air quality in other places and conditions.

4 CONCLUSIONS

To assess the PM distributions in underground subway environments, we conducted intensive measurements of PM in a subway station in Seoul, Korea, for three days. The concentrations averaged 59 μg m^{-3}, 37 μg m^{-3}, 111 μg m^{-3}, and 369 μg m^{-3} for the PM_{10} and 43 μg m^{-3}, 28 μg m^{-3}, 58 μg m^{-3}, and 132 μg m^{-3} for the $PM_{2.5}$ at the outdoor air inlet, in the concourse, on the platform, and in the tunnel, respectively. Strong temporal correlations between the levels measured at the different locations suggested extensive air exchange between the various subway sectors. In order to quantify the air flow exchange rates and PM fluxes between these areas, we applied a simple mass balance model using the $PM_{2.5}$-bound Fe as a tracer. The model validation revealed relative differences of less than 11% in the predicted PM_{10} and $PM_{2.5}$ temporal variations, confirming the accuracy of the simulations.

Furthermore, the model results indicated that 94% of the air mass in the concourse originated outdoors, arriving in the filtered air, whereas only 6% emanated from the platform. Compared to the concourse, the contribution of outdoor air to the air mass on the platform was relatively low (74%) despite the outdoor ventilation rate for this area being higher (by 46%). Additionally, the outdoors accounted for 84% and 87% of the PM_{10} and $PM_{2.5}$, respectively, in the concourse, but the tunnel accounted for 78% and 62% of the PM_{10} and $PM_{2.5}$ on the platform. Although the PM values inside the station met the Korean air quality guideline, those on the platform occasionally exceeded the recommended level. Hence, additional control strategies to improve the air quality on subway platforms must be implemented, with the emission reduction of tunnel-based sources—the most effective method, according to our study—receiving priority over measures such as enhancing the outdoor ventilation.
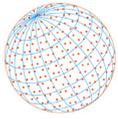
Finally, in our analysis, we assumed that the flow rates and PM concentrations remained steady for one hour. However, because trains pass every few minutes, these values may vary significantly on far shorter timescales. Also, the mass balance model should be verified for a longer sampling period. Nevertheless, our approach to estimating the air quality in subway spaces can be adapted to identify efficient mitigation measures for reducing pollution in other environments.

ACKNOWLEDGMENTS

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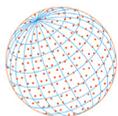
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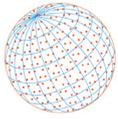
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