

PM_{2.5} Concentration and Composition in Subway Systems in the Northeastern United States

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OBJECTIVES: The goals of this study were to assess the air quality in subway systems in the northeastern United States and estimate the health risks for transit workers and commuters.

METHODS: We report real-time and gravimetric PM_{2.5} concentrations and particle composition from area samples collected in the subways of Philadelphia, Pennsylvania; Boston, Massachusetts; New York City, New York/New Jersey (NYC/NJ); and Washington, District of Columbia. A total of 71 stations across 12 transit lines were monitored during morning and evening rush hours.

RESULTS: We observed variable and high PM_{2.5} concentrations for on-train and on-platform measurements during morning (from 0600 hours to 1000 hours) and evening (from 1500 hours to 1900 hours) rush hour across cities. Mean real-time PM_{2.5} concentrations in underground stations were 779 ± 249 , 548 ± 207 , 341 ± 147 , 327 ± 136 , and $112 \pm 46.7 \mu\text{g}/\text{m}^3$ for the PATH-NYC/NJ; MTA-NYC; Washington, DC; Boston; and Philadelphia transit systems, respectively. In contrast, the mean real-time ambient PM_{2.5} concentration taken above ground outside the subway stations of PATH-NYC/NJ; MTA-NYC; Washington, DC; Boston; and Philadelphia were 20.8 ± 9.3 , 24.1 ± 9.3 , 12.01 ± 7.8 , 10.0 ± 2.7 , and $12.6 \pm 12.6 \mu\text{g}/\text{m}^3$, respectively. Stations serviced by the PATH-NYC/NJ system had the highest mean gravimetric PM_{2.5} concentration, $1,020 \mu\text{g}/\text{m}^3$, ever reported for a subway system, including two 1-h gravimetric PM_{2.5} values of approximately $1,700 \mu\text{g}/\text{m}^3$ during rush hour at one PATH-NYC/NJ subway station. Iron and total carbon accounted for approximately 80% of the PM_{2.5} mass in a targeted subset of systems and stations.

DISCUSSION: Our results document that there is an elevation in the PM_{2.5} concentrations across subway systems in the major urban centers of Northeastern United States during rush hours. Concentrations in some subway stations suggest that transit workers and commuters may be at increased risk according to U.S. federal environmental and occupational guidelines, depending on duration of exposure. This concern is highest for the PM_{2.5} concentrations encountered in the PATH-NYC/NJ transit system. Further research is urgently needed to identify the sources of PM_{2.5} and factors that contribute to high levels in individual stations and lines and to assess their potential health impacts on workers and/or commuters. <https://doi.org/10.1289/EHP7202>

Introduction

Subway systems are the veins and arteries of cities, moving people where they need to go. Their speed, accessibility, and affordability offer an alternative to often chaotic city streets. Globally, in 2017, approximately 168 million people used a metropolitan train daily (UITP 2018). Given the great potential for metropolitan subway systems to benefit urban communities, further growth in subway systems is anticipated. For example, studies in China have predicted opening new subway lines will decrease air pollution and provide substantial health benefits (Lu et al. 2018; Li et al. 2019).

Concerns exist, however, that air pollution exposures unique to underground subway systems occur during commuting. Previous studies have described airborne particulate matter (PM) concentrations within subways that are several times higher than PM levels in ambient air (Aarnio et al. 2005; Grass et al. 2010; Martins et al. 2016; Vilcassim, 2014). Although subway maintenance trains are usually diesel powered, passenger subway trains are powered by electricity, and therefore, one would not expect to have significant combustion-related sources in subway systems. Heavy metals have been shown to be major constituents of

airborne subway PM (Chillrud et al. 2004; Loxham et al. 2013; Lu et al. 2015a). Furthermore, Cha et al. (2016) demonstrated that PM_{2.5} (PM with aerodynamic diameter $<2.5 \mu\text{m}$) containing iron, copper, and manganese is generated by the interaction of a train's electrical current collector shoe with the supply rail. Sources other than friction of brakes and brushing of wheels may be important as well, particularly in busy subways where heavy maintenance machinery operate at night.

Commuters inhale subway-specific aerosols as they wait on a station platform and when riding the subway train. Transit system personnel are exposed for even longer periods of time while at work. Because numerous studies have shown that exposure to high concentrations of ambient PM_{2.5} is detrimental to human health (Kim et al. 2015; Lu et al. 2015b; Pun et al. 2017; Yang et al. 2020), there is justifiable concern about the potential for adverse health effects associated with the relatively poor air quality encountered in underground subway systems. Both daily commuters and transit workers may be at increased risk for cardiopulmonary disease if the integrated exposure levels approach or exceed the U.S. Environmental Protection Agency (U.S. EPA) ambient annual PM_{2.5} standard of $12 \mu\text{g}/\text{m}^3$ (U.S. EPA 2020a) or the American Conference of Governmental Industrial Hygienists' (ACGIH's) respirable (PM_{4.0}) occupational exposure guideline of $3 \text{mg}/\text{m}^3$ (Hearl 1998). Limited knowledge exists, however, regarding whether aerosol concentrations are elevated across U.S. subway systems, or whether the extent and composition of the aerosols vary from city to city, which would inform health risk estimates and abatement efforts.

This study builds on the work of Vilcassim et al. (2014), which measured PM_{2.5} and black carbon (BC) concentrations in the New York City (NYC) subway system. Here, we replicated and extended these findings by assessing air quality in NYC's Metropolitan Transit Authority (MTA) subway stations, the NYC/New Jersey Port Authority Trans-Hudson (PATH) stations

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Supplemental Material is available online (<https://doi.org/10.1289/EHP7202>).

The authors declare they have no actual or potential competing financial interests.

Received 7 April 2020; Revised 4 January 2021; Accepted 7 January 2021; Published 10 February 2021.

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and trains, and the Long Island Rail Road (LIRR). We compared the air quality data in these NYC area rail systems with air pollution concentrations and particle composition in the subway systems of Boston, Massachusetts; Philadelphia, Pennsylvania; and Washington, DC.

Methods

Sampling Sites

Real-time and filter-based concentrations of airborne particles were collected by study investigators from subway systems serving 4 metropolitan areas in the northeastern United States: NYC, Boston, Philadelphia, and Washington, DC. Data for NYC were collected from the NYC-MTA system in NYC and from two transit systems serving commuters to and from NYC: the PATH system serving the NYC borough of Manhattan and nearby portions of New Jersey, and the LIRR, a mostly aboveground heavy-rail train system that services Long Island, NY, including the NYC boroughs of Brooklyn and Queens, and Penn Station in Manhattan.

For sampling at each subway system, we selected one or more specific transit lines within the system, a number of stations along each line, and aboveground sites for ambient sample collection. Because of the diversity in the subway systems within the four targeted cities, a number of methods were used to select the sampling sites. NYC's MTA system has 424 subway stations, so we chose stations with known elevated $PM_{2.5}$ levels based on our previous study (Vilcassim 2014), where we measured $PM_{2.5}$ on 35 platforms in 30 subway stations along 5 MTA subway lines. In Boston and Washington, DC, subway lines and stations were selected based in part on ridership (with a goal of selecting stations with relatively high ridership in each system), along with other factors such as the number of underground stations and the delivery of electricity to the train (i.e., overhead and third rail). Philadelphia's subway system is relatively small and therefore the majority of stations were sampled.

Real-time measurements were performed during morning [from 0600 hours to 1000 hours (6:00–10:00 A.M.)] and evening [1500 to 1900 hours (3:00–7:00 P.M.)] rush-hour periods, except in Philadelphia where samples were collected during the evening rush hours only. Gravimetric measurements were also performed simultaneously with real-time measurements during morning and evening rush hours at a subset of stations with the highest real-time $PM_{2.5}$ concentrations in each system, excluding MTA-NYC and LIRR. A summary of sampling information for each system and transit line is provided in Table S1.

Real-Time Measurements

Real-time data were collected to provide a wide scale survey of the air pollution across many stations in multiple subway systems in a timely and efficient manner. Furthermore, real-time measurements were used to inform which sites were to be selected for gravimetric analysis (see "Gravimetric measurements" section below).

Real-time $BC_{2.5}$ and $PM_{2.5}$ concentrations were measured using a microaethalometer (AE 51; Magee Scientific/Aeth Labs) and a nephelometric-based DataRAM (pDR 1500, Thermo Fisher Scientific), respectively. These instruments were operated with 2.5 μ m cut point inlet cyclones and at 1-min averaging intervals. The real-time data were downloaded from the DataRAM and microaethalometer using pDR Port (Thermo Fisher) and microAethCom (version 2.2.4.0; Aethlabs) software, respectively.

For each transit system, real-time data were collected on the selected aboveground or underground subway platforms for 5 to 10 min before reboarding a train and moving on to the next stop.

Real-time ambient $PM_{2.5}$ and $BC_{2.5}$ concentrations were measured, also 5–10 min, approximately 0–8 m from an aboveground station entrance. On-train measurements were collected as each investigator rode on the train-car between stops. The on-train $PM_{2.5}$ concentration for each line was averaged for all interstation measurements that were 3 min or longer. The on-train values for LIRR were calculated as the average $PM_{2.5}$ concentration as the investigator rode approximately 30 min from Seaford station to Penn Station, and vice versa (3 runs in each direction).

To standardize real-time and gravimetric measurements across all subway systems, the real-time PDRs were zeroed with HEPA-filtered air before each run and set to report 1-min averages of $PM_{2.5}$. In addition to routine factory calibrations, all PDR real-time data were corrected with correction coefficients derived from calibration with gravimetric $PM_{2.5}$ concentrations (see details below).

Gravimetric Measurements

After review of the initial real-time $PM_{2.5}$ data for each line, 30–60 min Teflon gravimetric and quartz filter samples and real-time $PM_{2.5}$ data were collected simultaneously at the stations with the highest real-time $PM_{2.5}$ concentrations ($n=4$ or more per transit system except NYC's LIRR and MTA; see Tables S2 and S3).

For gravimetric measurement and carbon analysis, respectively, 37 mm diameter Teflon and prebaked quartz (Pall) filter samples were collected using a 2.5 μ m cut Personal Environmental Monitor (PEM) (SKC, Inc.), and a calibrated Leland Legacy Pump (SKC, Inc.) operated at 10 L/min. TeflonTM filters were preconditioned to U.S. EPA-recommended relative humidity (RH) and temperature for a minimum of 24 h, and the $PM_{2.5}$ mass concentration was calculated through standard gravimetric analysis using a microbalance (Model MT5, 1 μ g readability; Mettler) performed in a temperature- and humidity-regulated weighing room ($21\pm1^\circ\text{C}$ and $40\pm5\%$ RH). Laboratory blank samples were used to correct for daily variation in the micro-balance analyses.

Speciation Analysis

To evaluate the source(s) of $PM_{2.5}$ in the subway stations, compositional analysis of $PM_{2.5}$ was conducted on a subset of filters collected in the PATH-NYC/NJ, Boston, and Washington, DC, transit systems (simultaneously collected Teflon and quartz filters were not collected in Philadelphia, MTA-NYC, and LIRR). The concentration of organic and elemental carbon (OC and EC, respectively) on the quartz filters was determined using a Sunset Labs OCEC Analyzer (Sunset Instruments, Inc.) and the NIOSH (2003) OC/EC 5,040 method with a manufacturer's limit of detection of 1.25 $\mu\text{g}/\text{m}^3$ total carbon (TC). Laboratory and field blank quartz filters were included in all analyses. Note that total carbon ($\text{TC} = \text{OC} + \text{EC}$) is the primary carbon output reported in this study, because the presence of a high level of iron content (a major fraction of airborne subway $PM_{2.5}$) on the subway sample filters interferes with the thermal ramp curve for OC and EC (personal communication with Dr. Steven Chillrud, Columbia University), thereby limiting the precision to discern the forms of carbon species.

Trace element concentrations were determined on the Teflon filters by energy dispersive X-ray fluorescence spectroscopy (XRF) (Epsilon 5 ED-XRF; PAN Analytical B.V.), as previously described (Khodeir et al. 2012). Blank field and lab filters and spiked standards were included in the XRF analyses to determine uncertainty levels for each trace element. Trace element concentrations less than 3 times their uncertainty values, as determined by the instrument, were defined as below the detection limit.

Additionally, concentrations were corrected by subtracting the mean blank values for each element.

Statistical analysis. Each real-time station data point was calculated as the mean of each 5–10 min platform sample, excluding the first and last minute (i.e., to eliminate carryover from the 1-min averaging time of the instruments). Similarly, the first and last minute of the ambient and on-train measurements were removed from the analyses. The real-time PM_{2.5} data were corrected [correction factor (CF)] with RH measurements, collected simultaneously in the field with the PM_{2.5} data using the pDR-1500, using the equation (developed by Chakrabarti et al. 2004):

RH-adjusted PM_{2.5} concentration =

$$\text{RH-unadjusted PM}_{2.5} \text{ concentration} / \text{CF},$$

where $\text{CF} = 1 + 0.25 \text{RH}^2 / (1 - \text{RH})$.

Finally, the real-time PM_{2.5} data were adjusted with a correction factor developed using the gravimetric mass concentrations derived from the Teflon™ filters that were collected simultaneously with the RH-adjusted real-time PM_{2.5} data acquisition (minimum of $n=4$ for each city; Table S3 and Figure S1). Multiple regression functions were compared for derivation of this factor, and a power function was selected that provided the highest R^2 fit to the data. The resulting calibration equation is:

$$\text{Gravimetric-adjusted real-time PM}_{2.5} \text{ concentration} = 5.13(\text{RH-adjusted PM}_{2.5} \text{ pDR concentration})^{0.8681} \text{ (Figure S1)}$$

Data are presented as means (+SD) and medians. Correlation analysis (Pearson's correlation coefficient) of real-time PM_{2.5}, and BC_{2.5} data was done in R (version 3.6.1; R Development Core Team).

Results

Real-Time PM_{2.5} and BC_{2.5} Data

Real-time measurements were conducted to assess spatial differences in air pollution in the targeted subway lines. All real-time PM_{2.5} concentrations listed below were adjusted for RH and corrected with gravimetric data. The highest mean underground real-time PM_{2.5} concentrations were observed in the PATH subway system (mean $779 \pm 242 \mu\text{g}/\text{m}^3$; $n=26$ samples from 8 stations). The highest real-time PM_{2.5} concentration was $1,499 \mu\text{g}/\text{m}^3$ at the Christopher Street station during a morning rush hour (Table S2). The next highest adjusted mean real-time PM_{2.5} concentration was recorded in the MTA-NYC underground stations ($547 \pm 207 \mu\text{g}/\text{m}^3$; 12 samples from 3 stations) followed by underground stations in Washington, DC ($341 \pm 147 \mu\text{g}/\text{m}^3$; 84

Table 1. Real-time measured PM_{2.5} and BC_{2.5} concentrations by location (i.e., underground and aboveground stations, on-train, and aboveground ambient) for each urban transit system. All data, except in Philadelphia (February and August 2015), were collected from June to September 2019. Underground data were collected for 5 to 10 min at underground stations with single or multiple platforms (the latter were counted as multiple locations), whereas aboveground data correspond to aboveground station platforms. Ambient location samples were collected aboveground outside station entrances. On-train samples represent the concentration measured inside a train car during the ride between stations (for interstation rides greater than 3 minutes) and averaged for each run. RH, PM_{2.5} and BC_{2.5} was collected simultaneously. Real-time PM_{2.5} concentrations were adjusted for RH and corrected with gravimetric data.

System and line	Number of locations	Number of samples	PM _{2.5} (µg/m ³)				Number of samples	BC _{2.5} (µg/m ³)				RH (%)
			Mean ± SD	Median	Min	Max		Mean ± SD	Median	Min	Max	Mean ± SD
Boston												
Underground	24	96	327 ± 136	306	48	638	78	12.2 ± 12.3	10.4	1.04	103	49.9 ± 10.4
Aboveground	3	12	14.2 ± 5.9	13.8	7.2	28.7	10	2.2 ± 1.3	1.9	0.2	5.1	42.8 ± 10.6
On-train	3	12	182 ± 59.8	191	768	267	12	3.5 ± 2.2	3.8	0	6.3	45.8 ± 9.3
Ambient	3	5	10.0 ± 2.7	10.6	7.2	13.8	5	0.7 ± 0.9	0.3	0	2.1	53.0 ± 12.6
PATH-NYC/NJ												
Underground	8	26	779 ± 242	714	392	1499	20	28.5 ± 10.2	25.3	17.1	50.8	58.8 ± 8.5
Aboveground	ND						ND					
On-train	2	6	449 ± 261	378	219	958	4	13.9 ± 3.3	14.7	9.2	16.9	54.8 ± 8.3
Ambient	3	3	20.8 ± 9.3	20.3	11.8	30.3	2	2.2 ± 1.9	2.2	0.9	3.5	43.2 ± 1.0
MTA-NYC												
Underground	3	12	547 ± 207	509	263	959	12	22.4 ± 7.5	23.7	10.9	33.7	59.8 ± 3.5
Aboveground	ND	ND					ND					
On-train	4	4	343 ± 147	279	250	563	4	19.2 ± 22.5	8.8	6.2	52.8	50.9 ± 3.1
Ambient	2	4	24.1 ± 5.9	22.1	19.5	32.6	4	1.8 ± 0.7	1.8	0.9	2.6	56.2 ± 6.8
LIRR												
Underground	1	4	91.2 ± 38.9	103	36.7	123	4	6.3 ± 3.8	7.8	0.7	9.2	53.8 ± 13.1
Aboveground	1	3	13.8 ± 9.6	10.2	6.5	24.6	3	2.3 ± 1.5	2.1	0.9	3.9	53.6 ± 7.2
On-train	1	6	11.6 ± 5.10	10.7	7.33	18.6	6	1.1 ± 1.0	0.7	0.3	2.9	47.9 ± 5.7
Ambient	ND	ND					ND					
Philadelphia												
Underground	14	28	112 ± 46.7	100	49.7	226.6	21	7.1 ± 5.1	6.2	0.9	19.0	43.9 ± 5.9
Aboveground	ND						ND					
On-train	1	3	55.7 ± 2.2	56.0	53.4	57.8	3	2.0 ± 1.1	1.4	1.3	3.3	37.0 ± 3.7
Ambient	7	7	12.6 ± 4.8	15.5	5.3	17.1	7	2.3 ± 0.8	2.4	1.1	3.3	52.8 ± 24.0
Washington, DC												
Underground	21	84	341 ± 147	315	91.7	720	84	5.5 ± 2.7	5.1	1.3	16.0	52.3 ± 8.1
Aboveground	3	12	14.8 ± 7.5	15.6	5.0	24.4	12	1.9 ± 1.0	1.8	0.1	3.4	58.6 ± 12.1
On-train	2	7	186 ± 33.5	190	144	239	7	2.2 ± 0.9	2.2	0.9	3.4	45.7 ± 7.6
Ambient	1	3	12.0 ± 7.8	9.1	6.13	20.8	3	1.2 ± 0.8	1.6	0.3	1.6	58.4 ± 9.6
All Systems												
Underground	71	250	362 ± 226	315	36.7	1499	219	11.1 ± 10.9	7.6	0.7	103	51.8 ± 9.7
Aboveground	7	27	14.4 ± 6.7	14.1	5.0	28.7	25	2.1 ± 1.1	1.9	0.1	5.1	51.0 ± 13.1
On-train	10	38	205 ± 179	194	5.9	958	36	5.6 ± 9.1	2.9	0	52.8	47.4 ± 8.4
Ambient	16	22	15.1 ± 7.5	14.7	5.3	32.6	21	1.6 ± 1.0	1.6	0	3.5	53.0 ± 13.2

Note: BC, black carbon; Max, maximum; Min, minimum; ND, not done; RH, relative humidity.

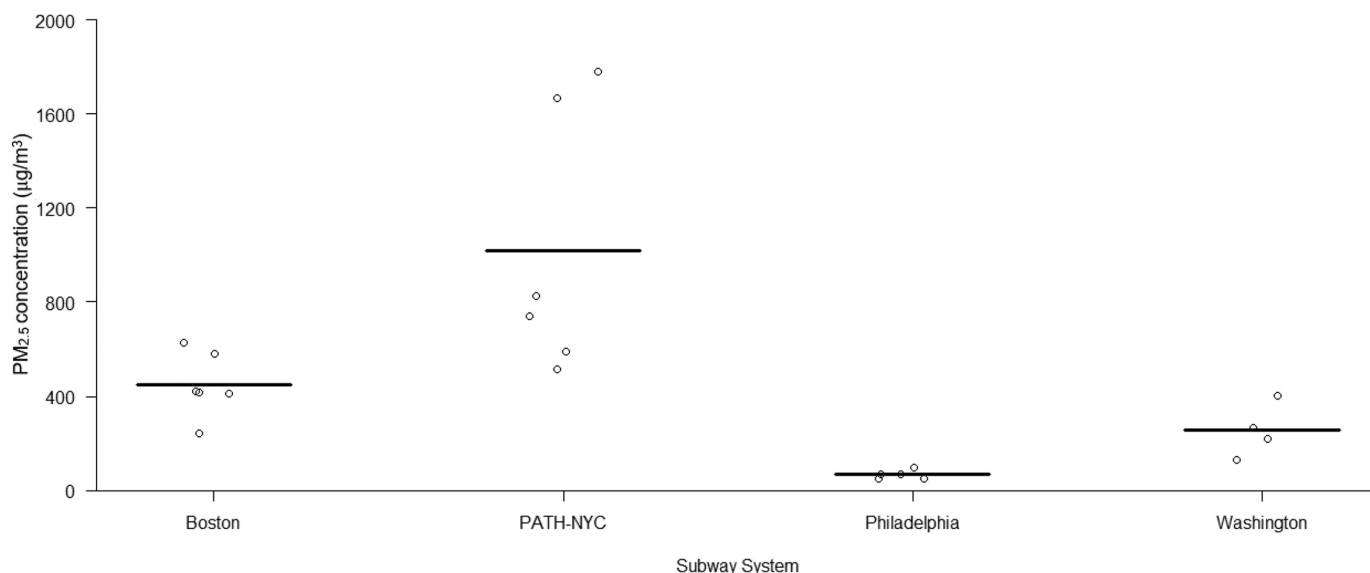


Figure 1. Mean (horizontal lines) gravimetric-based PM_{2.5} concentrations on underground subway platforms in four urban transit systems. The open circles represent the individual measurements. Stations represented in the Boston transit system: Broadway ($n=2$), Government Center–Blue Line ($n=2$), and Tufts Medical Center ($n=2$); in the PATH-NYC/NJ transit system: Newport ($n=2$) and Christopher Street ($n=4$); in the Philadelphia transit system: Race/Vine ($n=1$), City Hall ($n=1$), 13th Street ($n=1$), 15th Street ($n=1$), and 30th Street ($n=1$); and in the Washington, DC, transit system: Van Ness ($n=2$) and Capitol South ($n=2$). Filtered-based PM_{2.5} was collected for 30 to 60 min at 10 L/min.

samples from 21 stations), Boston ($327 \pm 136 \mu\text{g}/\text{m}^3$; 96 samples from 24 stations), Philadelphia ($112 \pm 46.7 \mu\text{g}/\text{m}^3$; 28 samples from 14 stations), and the LIRR ($91.2 \pm 38.9 \mu\text{g}/\text{m}^3$; 4 samples from 1 underground station) (Table 1). Real-time PM_{2.5} concentrations measured on trains (overall mean of $205 \pm 179 \mu\text{g}/\text{m}^3$) were consistently lower than corresponding concentrations in underground stations (overall mean of $362 \pm 226 \mu\text{g}/\text{m}^3$) (Table 1) but still substantially higher than concentrations in aboveground stations. Real-time PM_{2.5} concentrations estimated in aboveground stations (overall mean of $14.4 \pm 6.7 \mu\text{g}/\text{m}^3$) were similar to ambient PM_{2.5} concentrations measured outside the stations (overall mean of $15.1 \pm 7.5 \mu\text{g}/\text{m}^3$), and both were substantially lower than on-train or underground station PM_{2.5} concentrations.

Some differences in the adjusted real-time PM_{2.5} concentrations were observed among lines within the same subway system (Figure S2); however, for the most part, the range of PM_{2.5} concentrations overlapped among lines within the same city. For example, the subway platform levels for the Orange ($n=48$) and Red ($n=36$) subway lines in Washington, DC, were similar, whereas in New York, the PATH-NYC/NJ 33rd Street subway line ($n=20$) had a much greater mean PM_{2.5} concentration than the World Trade Center line ($n=6$) and the LIRR ($n=4$). In general, mean adjusted real-time PM_{2.5} concentrations for morning rush-hour samples were somewhat higher than mean concentrations for evening samples for most systems (Table S4).

As with the real-time PM_{2.5} measurements, the PATH-NYC/NJ system had the highest real-time BC_{2.5} concentrations in underground stations ($28.5 \pm 10.2 \mu\text{g}/\text{m}^3$). This level was followed closely by MTA-NYC ($22.4 \pm 7.5 \mu\text{g}/\text{m}^3$). There was considerable overlap in BC_{2.5} concentrations among the Boston ($12.2 \pm 12.3 \mu\text{g}/\text{m}^3$), Washington, DC ($5.5 \pm 2.7 \mu\text{g}/\text{m}^3$), LIRR ($6.4 \pm 3.8 \mu\text{g}/\text{m}^3$), and Philadelphia ($7.1 \pm 5.1 \mu\text{g}/\text{m}^3$) underground subway stations. As with PM_{2.5} concentrations, the overall mean underground subway platform BC_{2.5} ($11.1 \pm 10.9 \mu\text{g}/\text{m}^3$) was greater than the overall on-train mean BC_{2.5} concentration ($5.6 \pm 9.2 \mu\text{g}/\text{m}^3$). Both of these measurements were multiple times higher than the mean overall ambient and aboveground station values (1.6 ± 1.0 and $2.1 \pm 1.1 \mu\text{g}/\text{m}^3$, respectively).

As expected, PM_{2.5} and BC_{2.5} concentrations in underground stations were generally positively correlated (Pearson's correlation coefficient was 0.6 based on 219 data points; Figure S3). Within transit systems, the correlation was strongest for PATH-NYC/NJ ($r=0.9$, 20 data points) and weakest for Boston ($r=0.1$, 78 data points) (Figure S3).

The mean (+SD) percent RH for all underground stations was $51.8\% \pm 9.7\%$. In general, the percent RH was similar among the six urban transit systems with a high (mean+SD) of $59.8\% \pm 3.5\%$ in the underground subway stations in NYC's MTA and a low of $43.9\% \pm 5.9\%$ in Philadelphia.

Gravimetric Data

The filter-based gravimetric PM_{2.5} concentrations were found to be greater than the uncorrected real-time light scattering measurements. Across transit systems, gravimetrically calculated PM_{2.5} concentrations in the subways were 1.5- to 4-fold greater than the light-scattering real-time method [Table S3 shows the gravimetric and simultaneous real-time mean PM_{2.5} concentrations for the urban subway systems in Washington, DC ($n=4$), PATH-NYC/NJ ($n=6$), Boston ($n=6$), and Philadelphia ($n=5$)]. The underground stations in the PATH-NYC/NJ system had the highest mean (SD) gravimetric PM_{2.5} concentration of $1,020$ (557) $\mu\text{g}/\text{m}^3$ with 1-h gravimetric measurements of $1,669$ and $1,778 \mu\text{g}/\text{m}^3$ that occurred during two morning rush hours (Figure 1). Comparatively, the particulate air pollution in Washington, DC; Boston; and Philadelphia subway stations was lower, with mean gravimetric PM_{2.5} concentrations of 255 ± 114 , 450 ± 138 , and $66.9 \pm 18.8 \mu\text{g}/\text{m}^3$, respectively.

PM_{2.5} Speciation

To identify the source of PM_{2.5} in the subway stations, a study of PM_{2.5} composition was conducted in the PATH-NYC/NJ (Christopher Street and Newport stations), Boston (Government Center–Blue Line and Broadway stations), and Washington, DC (Van Ness and Capitol South stations) transit systems. Figure 2 shows the contribution of total carbon compounds and trace elements to total underground PM_{2.5} mass as measured as the total weight on the Teflon™ filter, aggregated across all stations. As

Table 2. Airborne Fe and Si aerosol concentrations in underground subway stations. The contribution of Fe and Si to the total PM_{2.5} mass was calculated using PM_{2.5} gravimetric data at 1 to 4 underground subway stations in three different urban systems.

System, line, and station	Number of samples	Si mean \pm SD ($\mu\text{g}/\text{m}^3$)	% contribution	Fe mean \pm SD ($\mu\text{g}/\text{m}^3$)	% contribution
Boston					
Government Center–Blue Line	1 ^a	4.5	1.9%	190	78.3%
Broadway	2	6.0 \pm 0.8	1.5%	162 \pm 14.6	39.1%
PATH-NYC/NJ					
Newport	2	42.2 \pm 18.6	7.5%	176 \pm 35.8	32.3%
Christopher Street	4	92.1 \pm 76.7	6.2%	329 \pm 116	29.7%
Washington, DC					
Van Ness	2	4.3 \pm 1.8	1.9%	180 \pm 44.7	76.5%
Capitol South	2	3.7 \pm 2.3	1.5%	141 \pm 81.1	56.7%

Note: Fe, iron; SD, standard deviation; Si, silicon.

^aOne sample was excluded due to a sampling error.

measured by XRF analyses, iron dominated the mean PM_{2.5} mass (Figure S4) and was $>100 \mu\text{g}/\text{m}^3$ (Table 2) at all sampled stations in Boston, NYC/NJ, and Washington, DC. Silicon was the next most abundant trace metal, making up an across-station average of approximately 3% of the PM_{2.5} mass, although its contribution to PM_{2.5} mass was greater in the PATH–NY/NJ stations (i.e., approximately 6% and 8% in the Christopher Street and Newport stations, respectively; Table 2). Other trace elements varied among the subway stations. For example, sodium, manganese, and barium were abundant in the Christopher Street station (PATH) and sodium, copper, and zinc predominated at the Capital South station in Washington, DC (Table S5). Table S6 displays the normalized elemental mass concentrations (μg element/mg PM_{2.5}) for other trace elements.

Total carbon species, which comprised 31% of PM_{2.5} mass, and estimated OC and EC concentrations and their contribution to total mass are presented in Table 3. The remainder of the mass, 6%, was not identified by the XRF and OC/EC analyses. As with trace elements, the contribution of TC to PM_{2.5} composition was found to vary among transit systems (Table 3 and Figure S4). Similar estimated airborne TC concentrations were observed in the targeted stations except in Government Center–Blue Line (Boston), where TC was 2 to 3 times higher than the other stations (Table 3). When these airborne TC concentrations ($\mu\text{g}/\text{m}^3$) were normalized as a ratio of μg OC/mg PM_{2.5}, particle composition differences were apparent among stations. For example, at the Christopher Street station in Manhattan, TC accounted for 3.7% of PM_{2.5} mass compared with 72.6% at the Government

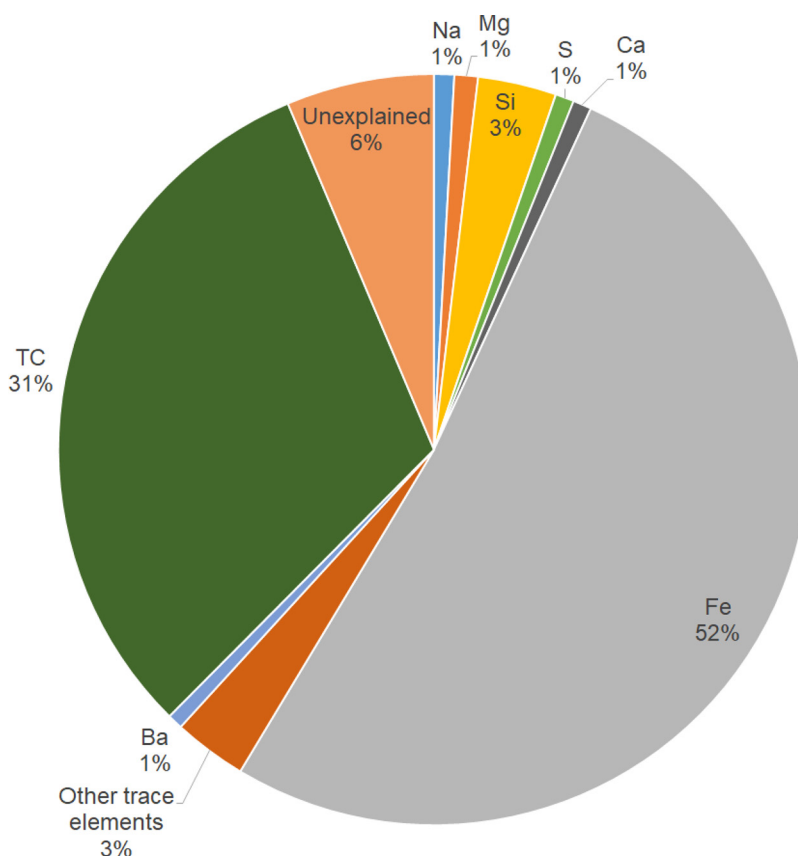


Figure 2. Average percent contribution of elemental constituents to total mass of PM_{2.5} collected on filters in underground stations in three urban transit systems. Each system is represented by the two stations with the highest measured PM_{2.5} concentrations: Government Center–Blue Line ($n=1$) and Broadway in Boston; Christopher Street ($n=4$) and Newport ($n=2$) in the PATH–NYC/NJ system; and Van Ness ($n=2$) and Capitol South ($n=2$) in Washington, DC. Trace elements analysis was conducted with XRF analysis of Teflon filters and carbon analysis was performed on the quartz filters using NIOSH's 5,040 method (NIOSH 2003). Note: Ba, Barium; Ca, Calcium; Fe, Iron; Mg, Magnesium; Na, Sodium; S, Sulfur; Si, Silicon; TC, total carbon.

Table 3. Airborne total, EC, OC carbon concentrations in underground subway stations. The contribution of carbon to the total PM_{2.5} mass was calculated using PM_{2.5} gravimetric data at one to two underground subway stations in three different urban systems.

System, Line, and Station		Total carbon		EC ^a		OC ^a	
		Mean ± SD (μg/m ³)	% contribution	Mean ± SD (μg/m ³)	% contribution	Mean ± SD (μg/m ³)	% contribution
Boston							
Government Center–Blue Line	1 ^b	177	72.6%	<LOD		177	72.6%
Broadway	2	70.6 ± 24.7	17.1%	12.2 ± 10.4	3.0%	58.4 ± 14.3	14.1%
PATH-NYC/NJ							
Newport	2	58.9 ± 29.8	10.9%	0.2 ± 0.01	0.03%	58.7 ± 29.8	10.6%
Christopher Street	2	57.2 ± 21.6	3.7%	<LOD		57.2 ± 21.6	3.7%
Washington, DC							
Van Ness	2	65.6 ± 15.7	27.0%	9.8 ± 2.0	4.0%	55.8 ± 13.7	23.0%
Capitol South	2	55.1 ± 6.2	20.7%	11.8 ± 5.2	4.4%	43.3 ± 1.0	16.3%

Note: EC, elemental carbon; LOD, limit of detection; OC, organic carbon; SD, standard deviation; TC, total carbon.

^aThe high iron content in subway PM_{2.5} samples interferes with the thermal ramp used in the analysis of EC and OC.

^bOne sample was excluded due to sampling error.

Center–Blue Line station in Boston. Despite the uncertainty associated with the measurement of OC and EC (due to the high iron content of subway PM_{2.5}), OC was present at far higher levels than EC (EC was present at low concentrations or below the detection limit in all stations).

Discussion

Our measurements and analyses reveal variable and, in places, very high PM_{2.5} exposures of commuters and transit workers in the underground subway systems of northeastern U.S. cities. The most extreme exposure, identified in a subway station on the PATH system (serving NJ and NYC), was higher than the previously published values for any subway station in the world (Martins et al. 2016; Moreno et al. 2017; Qiu et al. 2017; Van Ryswyk et al. 2017; Xu and Hao 2017; Lee et al. 2018; Minguillón et al. 2018; Mohsen et al. 2018; Moreno and de Miguel 2018; Choi et al. 2019; Loxham and Nieuwenhuijsen 2019; Pan et al. 2019; Shen and Gao 2019; Velasco et al. 2019; Smith et al. 2020), with a mean gravimetric PM_{2.5} concentration greater than 1,000 μg/m³ PM_{2.5} (Figure 1). The MTA-served subway stations in Manhattan also had poor air quality, with an adjusted real-time mean ± SD PM_{2.5} concentration of 548 ± 207 μg/m³.

Our particle measurements were similar to those measured previously in the MTA-NYC stations with high PM_{2.5} levels (Vilcassim et al. 2014) and much greater than aboveground ambient PM_{2.5} levels [it must be noted that the MTA-NYC subway stations monitored in the present study were a biased sample and chosen based on the high PM_{2.5} levels in the Vilcassim study (2014)]. Thus, during rush hour, the underground subway stations targeted in the NYC/NJ's MTA and PATH subway systems had significantly worse air quality, in terms of PM_{2.5}, than the targeted subway stations in Boston, Philadelphia, and Washington, DC. Philadelphia's subway stations, for example, had better air quality, although the mean real-time PM_{2.5} concentration was still several fold greater than the mean ambient PM_{2.5} concentration measured outside the Philadelphia subway stations. In addition, we cannot rule out spurious differences due to uncontrolled sources of variation related to sampling. However, our findings clearly indicate that PM_{2.5} concentrations in underground stations and measured on subway trains are much greater than aboveground ambient PM_{2.5} levels, at least during rush hour periods. In addition, we measured extremely high concentrations in individual underground stations in the MTA (NYC) and PATH (NYC/NJ) subway systems that, even if they represent extreme levels for these stations, raise serious health concerns and warrant additional investigation. In addition, underground PM_{2.5} concentrations were consistently higher than mean ambient PM_{2.5}

concentrations. Thus, our findings suggest that, at least in the northeastern U.S. transit systems included in our study, commuters are exposed to poor air quality during their time spent in underground subway stations. Moreover, exposures in at least some underground stations may be high enough to increase the risk of the adverse health effects associated with PM_{2.5}, even if they occur for relatively short periods of time.

It should be noted that most subway air pollution studies have relied on real-time data collected with light scattering instruments (Xu and Hao 2017) that have been factory calibrated, in the traditional manner, with Arizona road-dust (Curtis et al. 2008; Wang et al. 2016). Despite their many advantages (e.g., real-time data, autocorrection for temperature and RH), the output of real-time PM_{2.5} instruments can be affected by particle composition, shape, and water content, all physical factors that will variably affect light scattering. In the present study, we compared real-time and gravimetric PM_{2.5} concentrations during simultaneous 30- to 60-min sampling sessions conducted in the targeted subway systems (except MTA-NYC or the LIRR) and found, overall, that gravimetric values were 2–4 times greater than what was measured with the real-time light scattering device. This ratio is much higher than what has been reported for other environments and dust types (Wu et al. 2005; Wang et al. 2016, 2018; Patts et al. 2019), and this difference is most likely due to the large (e.g., as high as 60% of the total PM_{2.5} mass) contribution of iron, a dense metal, to the airborne PM_{2.5} in the targeted subway systems. Therefore, we adjusted our real-time PM_{2.5} data with a correction factor. Thus, this real-time/gravimetric ratio issue should be considered when interpreting health risks using published data from air quality studies of subway systems conducted throughout the world. Note that most of the samples collected at underground stations in the present study were selected because they had the highest estimated real-time PM_{2.5} concentrations in each system.

One of the highest unadjusted real-time mean subway system concentrations previously reported was 265 μg/m³ in Suzhou, China (Cao et al. 2017), whereas Seaton et al. (2005) and Smith et al. (2020) observed real-time, dust-type calibrated PM_{2.5} concentrations in a few stations in London, UK, that approached what was observed in PATH stations (Table S2), with a maximum 30-min mean concentration of 480 μg/m³ at one London station. Notably, Smith et al. (2020) observed a single 1-min peak of 885 μg/m³. The high pollution levels measured in London's subways did not reach the upper range of the PM_{2.5} levels in the PATH subway stations and particularly in the Christopher Street Station, which had a maximum 1-h gravimetric PM_{2.5} concentration of 1,780 μg/m³ during rush hour. The gravimetric PM_{2.5} concentrations measured at Newport Station, however, were more consistent with the peak values estimated in Smith et al. (2020). Comparison of our underground and ambient

PM_{2.5} data strongly suggests that ambient PM_{2.5} is not a likely source of the high PM_{2.5} levels observed in NYC's underground subway stations and that other sources such as the continual grinding of the train wheels against the rails, the electricity-collecting shoes, and diesel soot emissions from maintenance locomotives are important sources.

The contribution of TC to the PM_{2.5} mass concentration varied considerably among the two underground stations sampled at each of three transit systems (Table 3). TC constituted 6% of the particle composition in the PATH-NYC/NJ stations, whereas it composed 39% and 22% of PM_{2.5} in Boston and Washington, DC, respectively (Figure S4). Even within a single urban transit system, the TC concentrations varied between stations as was observed in Boston's Government Center–Blue Line (177 µg/m³) and Broadway stations (70.6 ± 24.7 µg/m³), albeit based on one and two samples, respectively. Broadway is an older station on the Red line, and Government Center is a much larger station with separate Blue and Green Line platforms and was renovated in the summer of 2016. Notably, TC, made primarily of the estimated OC component, dominated the Government Center–Blue Line aerosol, although the significance of this is unclear and further investigation into the sources of PM_{2.5} and the role of the mechanical design (e.g., ventilation) of each station is needed. Notably, there was relatively little EC (or the roughly equivalent BC_{2.5}) present in any of the six underground subway stations, an unexpected finding given the emphasis that multiple papers (Vilcassim et al. 2014; Choi et al. 2019) have placed on inorganic carbon species. A plausible source of EC would be diesel combustion in subway systems, for example, from diesel maintenance trains that operate in the MTA system. However, these trains are typically active only at night, and therefore their contribution to the composition of subway PM_{2.5} is unclear.

Iron accounted for the largest fraction of PM_{2.5} in the targeted subway stations, and frictional forces between the train wheels and rails and collection shoes and the third rail may account for this finding. The relative concentration of other elements was observed to vary among subway systems, suggesting that other sources (e.g., silicon as a marker for crustal material; arsenic as a marker for rodenticides) contribute to the airborne particles encountered by transit workers and commuters in subway stations. A previous report on PM composition in MTA stations in Manhattan agrees with the present findings. Chillrud et al. (2004) found similar ratios of iron/manganese, and chromium/manganese concentrations (i.e., components of different grades of steel), although some of the trace element concentrations in the present study are many times higher than those reported by Chillrud et al. Although other studies have documented low concentrations of noniron and carbon elements in subways (Minguillón et al. 2018; Lee et al. 2018), results in Shanghai, China, found that aluminum, silicon, and calcium made up more than 30% of PM_{2.5} (Lu et al. 2015a), suggesting that ambient soil particles can contribute to subway PM. Similarly, in Beijing subways, the iron concentration was outweighed by aluminum, potassium, sodium, calcium, magnesium, zinc, and barium (Pan et al. 2019). Thus, significant differences in PM composition exist among the underground subway systems across the globe, and it is likely that these differences are a result of source contributors that vary among systems.

Our results demonstrate considerable variability regarding the air quality that transit workers and commuters may encounter in the subway stations of the major cities in northeastern United States. Not only does the PM_{2.5} concentration vary among stations and cities, but the elemental composition of PM_{2.5}. Previous studies have demonstrated that underground depth (Vilcassim et al. 2014; Figueroa-Lara et al. 2019), station volume, age (Van Ryswyk et al. 2017), and ventilation (Martins et al. 2016) all

affect aerosol loading. Therefore, these subway system- and station-dependent differences were not unexpected in the present study. It is interesting to note that Martins et al. showed that more recently built stations do not necessarily have better air quality: The stations established in 2002 and 2009 in Oporto, Portugal, and Athens, Greece, had higher PM_{2.5} aerosol concentrations than a station built in 1983 in Barcelona, Spain. Nevertheless, there is evidence that common methods of reducing airborne PM are effective (Park et al. 2019), such as cleaning stations more often (Chen et al. 2017), improving ventilation (Moreno et al. 2014, 2017), using particle removal systems, and installing shields that confine track-generated particles from the boarding passengers (Guo et al. 2014).

Study Limitations

In the present study, there were relatively few interline differences in air quality among the subway lines within each city (Figure S2 and Table S1). One exception was the Blue line, which exhibited the highest PM_{2.5} concentration of the three targeted Boston subway lines. Because the Blue line is the most recently built of the three targeted Boston subway lines and would presumably have the best ventilation design (based on our subjective observations), this finding was unexpected. A limitation of this observation, however, is that we sampled air quality in two Blue line stations that were high train-traffic areas. A similar intrasubway system difference was, however, also observed in NYC/NJ's PATH system, where the mean PM_{2.5} level on the 33rd Street line was significantly greater than the World Trade Center line serviced by the PATH. As noted above, we did not collect information on all potential factors that might explain differences in air quality among different stations, subway lines, or transit systems.

We compared several transit systems using data collected at similar times of the day and generally within the same season. Thus, we have not cataloged the total potential variation of PM_{2.5} in each system. In particular, we sampled during a small number of days in Boston and Washington, DC, and our data generally represent only summer conditions. Thus, we do not know if the subway air quality changes significantly over season or time, although Van Ryswyk et al. (2017) have shown that the Toronto, Ontario, Canada, metro had higher PM_{2.5} concentrations than Montréal, Quebec, regardless of season. Another study limitation is that our PM_{2.5} and BC_{2.5} sample sizes for each station were relatively small ($n = 4$ for most stations). Although this study design allowed us to compare subway systems, we lacked sufficient power to compare PM_{2.5} concentrations among individual station platforms within a city's transit system. Nonetheless, certain stations were clearly more polluted than others. PM_{2.5} concentrations in the underground Christopher Street (PATH-NYC/NJ) were the highest among the 71 northeastern U.S. underground stations included in our study, and to our knowledge, were higher than any levels reported for any subway system across the globe (Martins et al. 2016; Moreno et al. 2017; Qiu et al. 2017; Van Ryswyk et al. 2017; Xu and Hao 2017; Lee et al. 2018; Minguillón et al. 2018; Mohsen et al. 2018; Moreno and de Miguel 2018; Choi et al. 2019; Loxham and Nieuwenhuijsen 2019; Pan et al. 2019; Shen and Gao 2019; Velasco et al. 2019; Smith et al. 2020).

In addition, it must be noted that the methodologies used to assess BC, OC, and EC were developed for measurement of PM composition collected under ambient outdoor conditions. The measurement of these carbon components was hampered, however, by the presence of large amounts, relative to ambient PM, of iron compounds. As demonstrated by other investigators, the dark color of some iron compounds can interfere with the reflectance measurement of BC, and the chemistry of the iron

compounds found in subway PM shifts the transition demarcation of OC and EC in the thermal ramp used by the Sunset Instrument analyzer. Therefore, we chose to present our data as TC (total carbon) concentrations to avoid the latter limitation. Regardless, the potential for underestimation of OC (i.e., caused by high levels of iron in PM_{2.5} collected on quartz filters in the subways; Figure S4) does not lessen the importance of OC as a major component of the PM_{2.5} collected in several subway stations.

Implications

The key issue with underground subway exposures is whether there is a significant increase in the risk for adverse cardiovascular and respiratory outcomes, given the well-documented association between PM_{2.5} and adverse health effects. With one notable exception, the PM_{2.5} concentrations measured in subway stations during morning and evening rush hours were generally 2 to 7 times the U.S. EPA's 24-h ambient air standard of 35 µg/m³. The one exceptional station (Christopher Street Station) on the PATH subway line connecting NJ to lower Manhattan had a maximum 1-h PM_{2.5} concentration of 1,780 µg/m³, with a mean gravimetric concentration of 1,254 µg/m³ (*n* = 4) (Table S3). If we assume that commuters are exposed to this level of PM_{2.5} for a typical 15-min total time (from/to home) spent on a subway platform and at 100 µg/m³ for two 20-min rides on the PATH subway trains each day, then a commuter's 24-h mean PM_{2.5} exposure concentration would increase from an assumed daily mean of 7.7 µg/m³ (for NYC metropolitan area; U.S. EPA 2020b) to 26.1 µg/m³. Given an association of a 6% increase in relative risk for each 10 µg/m³ increase in long-term (e.g., annual average) PM_{2.5} (Pope et al. 2004), this exposure scenario suggests that a typical commuter would be at an 11% increase in risk for cardiovascular mortality. However, this calculation assumes that the toxicity of underground subway PM_{2.5} is similar to that of ambient PM, which is uncertain in the absence of much-needed subway–health studies. It must be emphasized that this increase in individual risk for daily commuters differs in comparison to that for transit workers who spend considerably longer periods of time on the subway platforms (e.g., 8-h work shifts). The impact of exposure on transit workers is unclear because although they would be exposed to significantly greater PM_{2.5} accumulated doses (i.e., increased exposure time and breathing rates), workers are often considered “healthy,” and the most relevant applicable occupational exposure guidelines are for larger-diameter respirable dust, defined as PM_{4.0} (OSHA's Permissible Exposure Limit of 5 mg/m³ and ACGIH's threshold limit value of 3 mg/m³). In conclusion, these findings of poor air quality in subway systems should prompt further investigation as to the levels, sources, composition, and human health effects of the PM_{2.5} pollution in subway systems. However, even in the absence of such data, the results of our study already indicate that the Precautionary Principal (Science for Environment Policy 2017) would call for mitigation efforts, such as improved ventilation to protect the tens of thousands of subway workers and millions of daily commuters from potentially unwarranted health risks.

Acknowledgments

This research was supported by National Institute of Health (NIH)-funded Center (National Institute of Environmental Health Sciences (NIEHS) P30 ES000260 and NIEHS P30 ES009089) and Training (NIEHS T32 ES007324) grants. The authors thank R. Peltier for supplying equipment used in our Boston sampling campaign and B. DiSacre, J. Levis, M. Wren, and P. Brutsche, L. Brutsche, and A. Brutsche for their sampling work in Philadelphia.

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