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Authors: Singh, Subraham, Johnson, Glen, and Kavouras, Ilias G

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Subraham Singh, Glen Johnson and Ilias G Kavouras

Department of Environmental, Occupational, and Geospatial Health Sciences, City University of New York Graduate School of Public Health and Health Policy, New York, NY, USA.

ABSTRACT: Declining ambient $PM_{2.5}$ concentrations have been attributed to fuel consumption standards and emission controls of secondary sulfate and nitrate aerosol precursors from transportation and industrial sectors. As a result, the relative contribution of $PM_{2.5}$ sources is modified, shifting $PM_{2.5}$ trends, physicochemical characteristics, and health effects. Carbonaceous fine aerosol account for most of $PM_{2.5}$ mass in the US. This study aims to examine the spatiotemporal trends of ambient $PM_{2.5}$ levels and their association with primary $PM_{2.5}$ emissions from anthropogenic activities and fires in the New York/New Jersey metropolitan statistical area (NYNJ MSA) airshed. $PM_{2.5}$ mass concentrations were obtained from the U.S. Environmental Protection Agency (USEPA) Air Data. Ambient $PM_{2.5}$ mass levels declined on average by 47%, at a rate of $-0.61 \pm 0.01\,\mu g/m^3$/year in urban locations and $-0.25 \pm 0.01\,\mu g/m^3$/year in upwind and peri-urban locations over the 2007 to 2017 period. The strong spatial gradient in 2007, with high $PM_{2.5}$ levels in urban locations and low $PM_{2.5}$ levels in peri-urban locations gradually weakened by 2013 but re-appeared in 2017. Over the same period, primary $PM_{2.5}$ emissions declined by 52% from transportation, 15% from industrial, and 8% from other anthropogenic sources corresponding to a decrease of 0.8, 0.9, and $0.6\,\mu g/m^3$ on ambient $PM_{2.5}$ mass, respectively. Wildland and prescribed fires emissions increased more than 3 times adding $0.8\,\mu g/m^3$ to ambient $PM_{2.5}$ mass. These results indicate that (i) fire emissions may impede the effectiveness of existing policies to improve air quality and (ii) the chemical content of $PM_{2.5}$ may be changing to an evolving mixture of aromatic and oxygenated organic species with differential toxicological responses as compared to inert ammonium sulfate and nitrate salts.

KEYWORDS: Fine particles, transportation, emission, air quality, megacity

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CORRESPONDING AUTHOR: Ilias G Kavouras, Department of Environmental, Occupational, and Geospatial Health Sciences, City University of New York Graduate School of Public Health and Health Policy, 55 West 125th Street, New York, NY 10027, USA. Email: ilias.kavouras@sph.cuny.edu

Introduction

Both short[1] and long-term[2] exposures to fine particulate matter ($PM_{2.5}$; particles with aerodynamic diameter [$d_a < 2.5\,\mu m$]) increase the relative risk of cardiovascular and pulmonary morbidity and mortality. Ecologically, ambient $PM_{2.5}$ impairs visibility, adversely impacts ecosystems, modifies infectious diseases distribution, and amplifies the magnitude and frequency of natural disasters.[3] $PM_{2.5}$ penetrates deeper into the lung's alveolar region, inducing inflammatory and oxidative stress responses that trigger or exacerbate a range of harmful health outcomes.[4,5] High rates of premature mortality, asthma attacks, chronic obstructive pulmonary disease (COPD), lung cancer among certain vulnerable groups, particularly underserved and under-resourced ethnically and racially diverse minorities, immunocompromised, elderly, and children who are exposed to elevated $PM_{2.5}$ concentrations were observed.[6-8]

$PM_{2.5}$ is composed of a mixture of chemical species depending on the type and intensity of sources. Primary sources include anthropogenic activities such as transportation, domestic heating, industrial activities, and to a lesser extent, long-range transport of windblown dust.[9-11] They are composed of heavy metals (Ni, V, Cr, An, Cu, Zn) and crystal elements (Al, Si, Ca, Fe, Ti), elemental carbon (EC) and semi- and non-volatile organic compounds. Ultrafine sulfate (SO_4^{2-}) and nitrate (NO_3^-) particles (with $d_a < 100\,nm$) are formed through the oxidation of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) emitted from fossil fuel combustion and subsequent neutralization by ammonia (NH_3). Reactions of biogenic hydrocarbons and anthropogenic chemical species with atmospheric oxidants also yield the formation of ultrafine secondary organic aerosol.[12] Wildfires, prescribed and agricultural fires, as well as wood combustion are increasingly important sources of $PM_{2.5}$.[13] Paved and unpaved road dust can account for up to one-third of $PM_{2.5}$ mass, particularly in arid urban environments.[14] Organic compounds, transition metals, elemental carbon, ions, viruses, bacteria, house allergens, spores, and pollen are often found in $PM_{2.5}$.[15,16]

Transportation has been recognized as an important source of $PM_{2.5}$ accounting for about 17% to 23% of urban $PM_{2.5}$.[11] Over the past decades, cleaner fuels and better engines for passenger cars, trucks, small engines, commercial marine vessels, and locomotives have been developed to reduce emissions. As a result, primary $PM_{2.5}$ emissions from transportation sources have been declining. More specifically, between 2008 and 2017, total transportation emissions of primary $PM_{2.5}$ declined by 48%, ranging from 45% for on-road and highway emissions to 46% for trains, ships, and locomotives emissions and up to 55% for off-road emissions in the United States.[17] Transportation NO_x and VOCs emissions also declined, yet, evolving atmospheric chemistry conditions may enhance $PM_{2.5}$ mass up to

Table 1. The characteristics (ID#, name, type, latitude, longitude, elevation, distance from the Division Street site [set as the reference site], and population within an 8-km radius of $PM_{2.5}$ monitoring sites in New York City metropolitan area).

ID#	SITE NAME	LATITUDE (°N)	LONGITUDE (°W)	ELEV.	DISTANCE FROM DIVISION STR. SITE (KM)	POPULATION (<8 KM)
1	Division street	40.71436	–73.99518	17	—	4 539 895
2	IS 45	40.79970	–73.93432	3	10.8	4 661 089
3	PS 19	40.73000	–73.98400	9	2.0	4 710 192
4	JHS 126	40.71961	–73.94771	6	4.1	5 172 079
5	Queens college	40.73614	–73.82153	25	15.0	2 825 439
6	Richmond post office	40.63307	–74.13719	16	14.7	1 221 806
7	Jersey city firehouse	40.72541	–74.05229	6	4.9	2 693 964
8	Fort Lee library	40.85226	–73.97331	91	15.7	3 238 133
9	Elizabeth lab	40.64144	–74.20836	5	19.7	1 163 841
10	Babylon	40.74529	–73.41919	27	48.6	671 479
11	Newburgh	41.49916	–74.00885	127	86.1	195 678
12	Paterson	40.91838	–74.16809	21	27.6	944 841
13	Chester	40.78763	–74.67630	278	57.9	205 712
14	Bridgeport	41.17083	–73.19472	7	84.6	382 968

0.9 μg/m^3 and ozone (O_3) up to 5 ppbv.[18] Hydroxyl (OH) radicals may be available to react with VOCs because of the declining levels of SO_2 and NO_x, leading to the formation of secondary organic aerosol and O_3.[19]

Ambient $PM_{2.5}$ mass concentrations in New York State including New York City declined up to 7 μg/m^3 at urban sites for the 2000 to 2015 period. These changes were greatly attributed to reduced local and transported of coal-fired power plants emissions and other fossil-fuel combustions processes, with emphasis on secondary sulfate and nitrate precursors, sulfur dioxide, and nitrogen oxides.[19-21] Very little attention has been paid to primary $PM_{2.5}$ emissions that contain hazardous carbonaceous aerosol including elemental carbon and a complex mixture of organic compounds such as polycyclic aromatic hydrocarbons (PAHs).[19] The New York/New Jersey Metropolitan Statistical Area (NY/NJ MSA) is the largest and denser urban agglomeration in North America, with almost 20 million people residing in a relatively small area (17 314 km^2), particularly those within New York City (NYC). The region meets the annual and daily 2012 National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ of 12 and 35 μg/m^3, but it is a non-attainment area for ozone. There were over 4 000 000 vehicles in NYC and adjacent Westchester and Long Island counties (Nassau and Suffolk), accounting for 41.6% of registered vehicles in New York state. Most of them (95.75%) were gasoline-powered, followed by diesel engines (3.25%).[22] $PM_{2.5}$ levels in eastern US were also significantly affected by wildfires during the 2018 Long Island Sound Tropospheric Ozone Study (LISTOS).[23] Increased $PM_{2.5}$, black carbon (BC), and biomass burning tracers in Long Island were associated with smoke plumes from distant fires. The aims of this study were (i) to characterize the spatial and temporal trends of $PM_{2.5}$ mass concentrations in the broader NY/NJ MSA airshed and (ii) to investigate the impact of primary $PM_{2.5}$ emissions from local transportation and biomass burning on $PM_{2.5}$ levels, using publicly available $PM_{2.5}$ measurements through the USEPA Air Data system.

Materials and Methods

Air pollution measurements

Daily $PM_{2.5}$ mass concentrations measured at 14 sites in the NYNJ MSA region for the 2007 to 2017 period were obtained from the USEPA Air Data system (Table 1).[24] The air quality monitoring sites, population, and major traffic corridors are presented in Figure 1. Six of the sites were located within NYC; Division Street (#1); and PS-19 (#3) in Lower Manhattan, and JHS in Brooklyn (#4) with a population ranging from 4.54 to 5.17 million people within 8-km radius. IS-45 (#2) in East Harlem with a population of 4.66 million people with an 8-km radius.[25] Queens College in Queens (#5), and Richmond Post Office (#6) with populations of 2.8 and 1.2 million people within 8-km. Two sites located outside of NYC were in Babylon (#10, ~650 000 people in 8-km radius) and Newburgh (#11, about ~200 000 residents within 8 km). Five sites were in New Jersey, 4 of them (#7, #8, #9, and #12) in densely populated

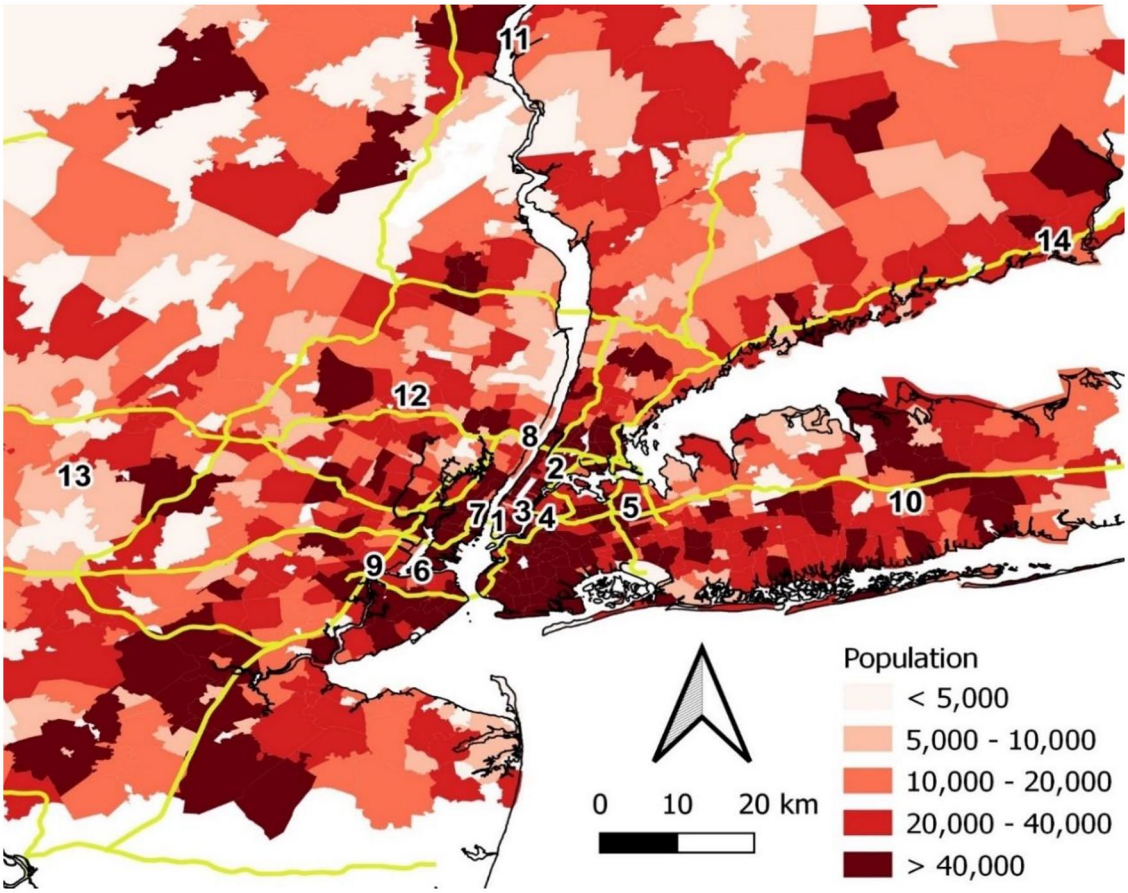

Figure 1. The locations of ambient $PM_{2.5}$ monitoring sites, 2019 population (by US Census tract) and primary road network in NYC metropolitan area (see Table 1 for site characteristics).

areas (from 900 000 to 3 300 000 people within 8-km radius), while the site at Chester was upwind of NYC (#13-200 000 people within 8-km). One site was located in Bridgeport, CT, (#14, Figure 1, with 400 000 people living within an 8-km radius). $PM_{2.5}$ mass was monitored daily at sites #1, #5, #7, #9, and #11 and in 1-in-3 days frequency at the remaining sites.

Emissions inventories

Annual primary $PM_{2.5}$ emissions from 2007 to 2017 for New York, New Jersey, and Connecticut were obtained from the USEPA National Emissions Inventory (NEI).[26] The emissions are reported in 15 Tier-1 categories (Supplemental Table S1) as follows: 1 to 2: highway and off-highway vehicles (2 groups), 3 to 4: prescribed and wildfires (2 groups), 5: chemical and applied product manufacturing, 6 to 8: fuel combustion by electrical utilities, industrial, and other activities (3 groups), 9: metals processing, 10: other industrial processes, 11: petroleum and related industries, 12: solvent utilization, 13: storage and transport, 14: waste disposal and recycling, and 15: miscellaneous. The NEI Tier files used to develop the national and state trends based on emissions inventories are for the years 2008, 2011, 2014, and 2017. On/off highway vehicular emissions were updated for 2007, 2009, and 2010. The 2015 and 2016 emissions were computed through interpolation of the 2017 emissions. Wildfires are included in Miscellaneous for 2007 and 2008. The 2008 wildfire emissions were used for 2009 and 2010, 2011 wildfire emissions for 2012 and 2013, and 2014 wildfire emissions for 2015 and 2016. The Tier 1 groups were combined in 4 sectors as follows: Transportation: categories 1 to 2; Fires: categories 3 to 4; Industrial: categories 5 to 13; and Other: categories 14 to 15. Note that wood and biomass combustion for industrial purposes is included in the fuel combustion Tier 1 categories. Annual $PM_{2.5}$ mass concentrations and emissions were tested for normality using the Shapiro-Wilk test. The Pearson correlation coefficient was assessed at $\alpha = .05$ by site to determine correlations between ambient and emitted $PM_{2.5}$.

Trend analysis

The monthly mean $PM_{2.5}$ mass was computed for months with more than 75% of scheduled $PM_{2.5}$ mass concentrations. The annual trend was computed by applying the non-parametric sequential Mann-Kendall test at a confidence level of 95%.[27] Analyses were done using SPSS (Version 26) (IBM Analytics, Armonk, NY). Two approaches were used to assess the spatial variability of $PM_{2.5}$ mass concentrations. First, the daily paired

absolute (ΔC) and the percent relative ($\%\Delta C/\text{Ref}$) $PM_{2.5}$ mass concentration differences and the coefficient of divergence (COD) were computed.[28] The Division Street location in Downtown Manhattan (Site #1 in Figure 1) was set as the reference site because of its central location to the study area. The paired ΔC and $\%\Delta C/C_{\text{Ref}}$ evaluate temporal correlations and systematic differences between the sites and site-to-site variation. COD assess the spatial uniformity of measurements with COD close to unity being indicative of spatial gradient. Secondly, the local Moran's I and its significance (using standardized Z-score) was computed to examine clustering of $PM_{2.5}$ mass concentrations to assess spatial heterogeneity using equations (1) and (2) below[29]:

$$I = \frac{n \cdot \sum_{i=1}^{n} \sum_{j=1}^{n} w_{ij} \cdot \left(x_i - \bar{x}\right) \cdot \left(x_j - \bar{x}\right)}{\sum_{j=1}^{n} \left(x_j - \bar{x}\right)^2} \quad (1)$$

and

$$Z = \frac{I - E(I)}{\sqrt{var(I)}} \quad (2)$$

x_i and x_j are the annual $PM_{2.5}$ mass concentration at ith and jth *sites* for i, j, $\bar{x}$ was the average $PM_{2.5}$ mass concentration in all sites, w_{ij} was the Euclidean distance between 2 sites, n is the number of sites, $E(I)$ is the mathematical expectation of local Moran's I and var(I) is the variance of the local Moran's I.

The spatial patterns of $PM_{2.5}$ mass are classified in 5 categories as follows: H-H ($I>0$ and $Z>0$) for spatial clusters with high $PM_{2.5}$ mass, L-L ($I>0$ and $Z<0$) for spatial clusters with low $PM_{2.5}$ mass, H-L ($I<0$ and $Z>0$) for spatial clusters with high $PM_{2.5}$ mass surrounded by low $PM_{2.5}$ mass clusters, L-H ($I<0$ and $Z<0$) for spatial clusters with low $PM_{2.5}$ mass surrounded by clusters of high $PM_{2.5}$ mass, and not significant, for no spatial clusters. For the $PM_{2.5}$ annual trends, considering that a declining annual trend was computed for all sites, the clusters were indicative of: H-H, spatial cluster with slowest $PM_{2.5}$ mass decline rate, L-L spatial clusters with the fastest $PM_{2.5}$ mass decline rate decline, H-L spatial clusters of slow $PM_{2.5}$ mass decline rate surrounded by clusters of fast $PM_{2.5}$ mass decline rate, L-H spatial clusters of fast $PM_{2.5}$ mass decline rate surrounded by clusters of slow $PM_{2.5}$ mass decline rate. Analysis was done using GeoDa (v. 1.14.0.24).

Results and Discussion

Temporal trends

Table 2 shows the 2017 annual mean and 2007 to 2017 annual trend $PM_{2.5}$ mass concentrations for each site. The monthly mean $PM_{2.5}$ mass concentration in NYC, NJ, and the peri-urban sites in NY, NJ, and CT are illustrated in Figure 2A to C. $PM_{2.5}$ mass concentrations were comparable among all sites in the study area (ranging from 5.8 to 9.6 μg/m³), substantially lower than the National Ambient Air Quality Standard (NAAQS) of 12 μg/m³. Most of the sites exhibited a clear seasonal profile with the higher $PM_{2.5}$ levels in the winter and summer (Figure 2). This pattern is consistent with the seasonal profiles of ammonium nitrate (high in winter) and ammonium sulfate (high in summer) in source apportionment and photochemical models in NYC.[19,20] Domestic woodburning emissions of primary $PM_{2.5}$ were more pronounced in winter. Secondary organic aerosol formation is negligible during the winter due to insufficient incoming solar radiation. Aged wildfires smoke, including both primary and secondary $PM_{2.5}$, and recreational fires were prevalent in the summer.[20]

Table 2. The 2017 $PM_{2.5}$ mass concentration (μg/m³) and 2007 to 2017 annual trend (μg/m³/y) (mean ± standard error).

SITE ID AND NAME	2017 MEAN $PM_{2.5}$	ANNUAL TREND
Division street	6.9 ± 0.1	−0.56 ± 0.01
IS 45	7.5 ± 0.1	−0.47 ± 0.01
PS 19	8.8 ± 0.2	−0.61 ± 0.01
JHS 126	7.6 ± 0.1	−0.44 ± 0.01
Queens college	7.1 ± 0.1	−0.39 ± 0.01
Richmond post office	7.0 ± 0.1	−0.46 ± 0.01
Jersey city firehouse	8.1 ± 0.1	−0.38 ± 0.01
Fort Lee library	7.2 ± 0.1	−0.34 ± 0.01
Elizabeth lab	9.6 ± 0.1	−0.34 ± 0.01
Babylon	6.7 ± 0.1	−0.33 ± 0.01
Newburgh	5.8 ± 0.2	−0.42 ± 0.01
Paterson	7.8 ± 0.2	−0.38 ± 0.01
Chester	6.0 ± 0.1	−0.25 ± 0.01
Bridgeport	6.9 ± 0.1	−0.30 ± 0.01

For all sites, $PM_{2.5}$ mass concentrations consistently declined during the 2007 to 2017, from −0.25 ± 0.01 μg/m³/year in upwind Chester to −0.61 ± 0.01 μg/m³/year to NYC (Site: PS 19) (Table 2), in agreement with previous estimates since 2000.[19] Regional sources of secondary inorganic species declined from about 50% (46%-57%) in 2002, to 25% to 46% of $PM_{2.5}$ in 2018 in NYC.[19] Changes in the sulfur content of fuel types (ie, Clean Heat: phasing out No. 4 and No. 6 fuel oils with high sulfur content) may have reduced primary $PM_{2.5}$ emissions from oil boilers.[19,30] The ambient levels of tracers of industrial emissions (Cr, Cu, Fe, and Mn) and crude oil (Ni, V) combustion also declined during the same period.[19,31] Slightly higher declining rates were computed for heavily populated urban sites in NYC and NJ as compared to those computed for peri-urban sites. A less pronounced decline has been also observed in the Upstate New York region farther away from

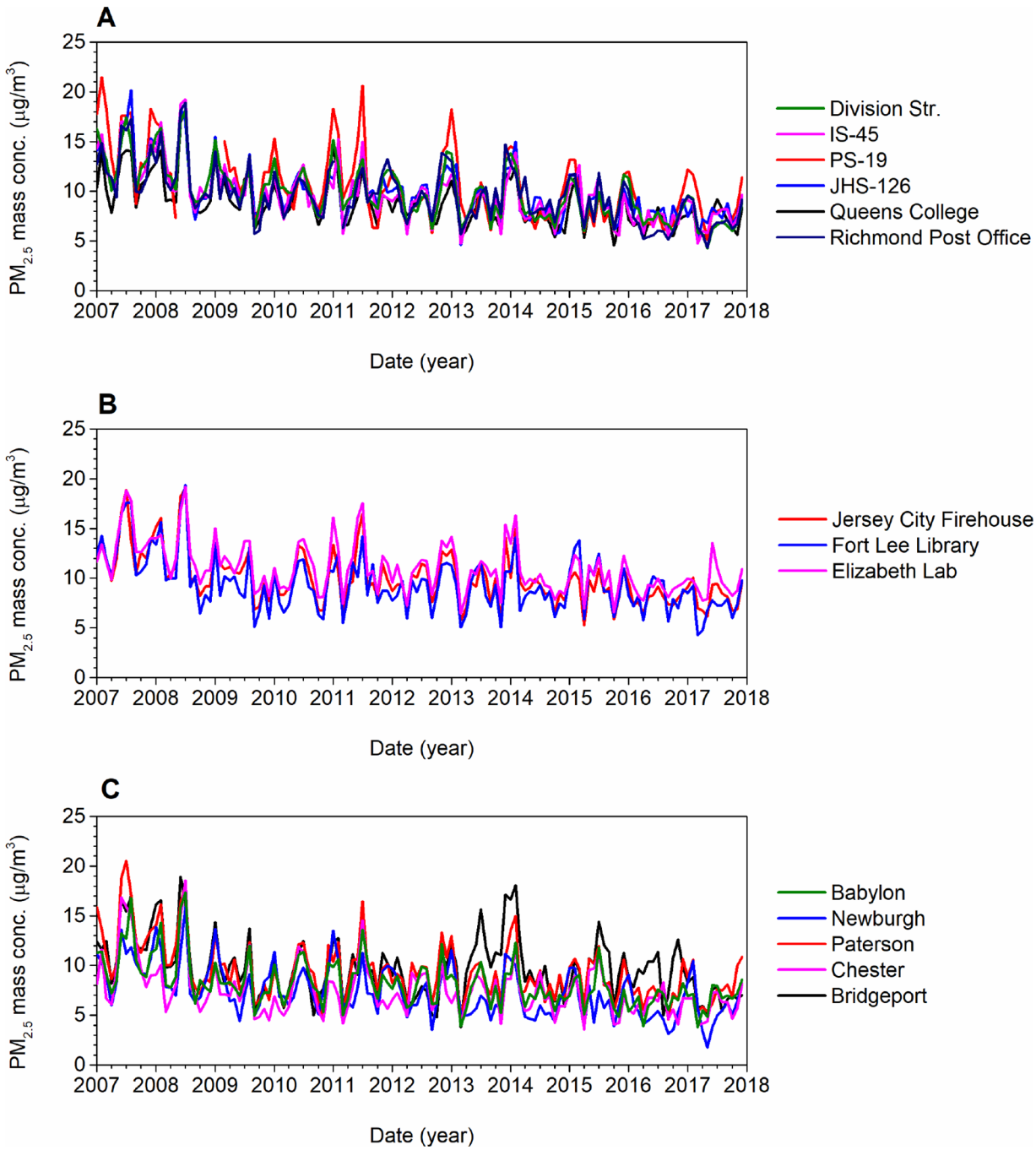

Figure 2. The mean monthly $PM_{2.5}$ concentrations at (A) urban sites within NYC, (B) urban sites and in NJ, and (C) peri-urban sites in NJ, NY, and CT.

the major urban centers with reduction rates of 3 to 4 μg/m^3/year.[19] The difference between the annual $PM_{2.5}$ declining rates in urban and peri urban site may be due to local primary $PM_{2.5}$ emissions controls from traffic and industrial process.

Spatial trend: Coefficient of divergence and Moran I spatial autocorrelation

The coefficient of divergence (COD), absolute (ΔC), and relative ($\%\Delta C/C_{Ref}$) (median and standard deviation [σ]) differences (compared to reference site [Division Street]) of $PM_{2.5}$ concentrations are shown in Table 3. The lack of a spatial pattern in urban sites (#2-9) as suggested by the low COD (from 0.18 to 0.22) may be due to the dominant contribution of regional aerosol. For the peri-urban sites (#10-14) located at farther distances from the reference site. COD values increased from 0.24 to 0.50, suggesting the existence of a stronger west-to-east spatial trend (COD values increase from 0 to 1 for spatial gradients).

The local Moran's *I* clusters for the 2007 to 2017 and $PM_{2.5}$ annual trends are illustrated in Figure 3A to L. "H-H" clusters were identified in highly populated urban areas demonstrating positive autocorrelation for spatial clusters of high $PM_{2.5}$ mass concentrations for the 2007 to 2012 period. The gradient

Table 3. The mean COD, absolute (ΔC), and relative ($\%\Delta C/C_{Ref}$) (median and standard deviation [σ]) differences (compared to Division street site) of $PM_{2.5}$ concentrations.

SITE ID AND NAME	COD	ΔC (MEDIAN [SD])	$\%\Delta C/C_{REF}$ (MEDIAN [SD])
Division street	—	—	—
IS 45	0.20	−0.30 (2.49)	−2.78 (116.43)
PS 19	0.18	0.50 (2.43)	5.87 (86.18)
JHS 126	0.21	0.05 (2.70)	0.00 (133.14)
Queens college	0.20	−1.00 (2.51)	−11.18 (72.3)
Richmond post office	0.22	0.00 (2.15)	0.00 (64.72)
Jersey city firehouse	0.19	0.20 (2.95)	1.81 (112.32)
Fort Lee library	0.22	−0.50 (3.32)	−6.21 (116.02)
Elizabeth lab	0.21	1.10 (3.33)	11.83 (150.22)
Babylon	0.24	−1.60 (3.37)	−17.89 (101.77)
Newburgh	0.36	−2.10 (2.96)	−26.83 (48.01)
Paterson	0.22	−0.60 (3.42)	−7.31 (97.59)
Chester	0.29	−2.30 (3.67)	−26.35 (91.37)
Bridgeport	0.50	0.00 (3.97)	−0.65 (102.82)

declines over time leading to the lack of spatial clustering for the 2013 to 2016 period. A weak "H-H" spatial clustering reappeared in 2017. No spatial clustering was observed for peri-urban sites. The spatial pattern changes over time were consistent with spatial correlation of the annual trends with "L-L" clusters being computed for the urban sites (note that low annual trends were indicative of rapid $PM_{2.5}$ mass concentration decline). Although the number of features used in this study (n = 14) was lower than the suggested features count (n = 30), the trends were consistent with those using site-specific absolute and relative concentration differences and COD.

The observed spatial trends may be due to the rapid decline of highly correlated $PM_{2.5}$ levels in urban areas as compared to those in peri-urban areas prior to 2012, because of emission controls. The re-appearance of spatial gradient in 2017 may be attributed to changes in local emissions and atmospheric chemistry including synergistic effects of organic carbon emitted from biomass burning.[20] It can be linked to the availability of atmospheric hydroxyl radicals due to reductions in SO_2 and NO_x emission.[19,32] It has been recognized that the relative abundance of organic carbon on $PM_{2.5}$ mass has increased.[33] The 2018 particulate organic carbon (OC) levels in NYC were up to 6% higher than those measured in 2002 levels with approximately half of that from upwind regional sources.[19] Secondary organic aerosol (SOA) formed from the photooxidation of freshly emitted anthropogenic and biogenic volatile organic compounds may account for up 64% of total OA in the study area.[34] The wide range of organic compounds, from long-chain aliphatic hydrocarbons, PAHs, and polyfunctional macromolecules presents a significant challenge to control ambient $PM_{2.5}$ mass concentrations.[36]

$PM_{2.5}$ emissions trends

Figure 4 illustrates (A) the relative contributions of primary $PM_{2.5}$ emissions from fires, traffic, industrial, and other sources in 2007 and 2017 and (B) the Pearson correlation coefficient between annual trends of ambient $PM_{2.5}$ measurements in each site and primary $PM_{2.5}$ emissions. The total primary $PM_{2.5}$ emissions merely declined by 2% between 2007 and 2017. Emissions from the transportation sector were reduced by 52%, accounting from 22% of the 2007 $PM_{2.5}$ emissions down to 11% of 2017 $PM_{2.5}$ emissions Industrial emissions reduced by 11%, accounting for 46% in 2007, and 42% in 2017. For $PM_{2.5}$ primary emissions from other sources were reduced by 20% without changes in the relative contribution of $PM_{2.5}$ emissions over time. The consistent declining trends of annual $PM_{2.5}$ levels and traffic, industrial and other primary $PM_{2.5}$ emissions was further corroborated by the strong R values (Figure 4B).

Primary $PM_{2.5}$ emissions from fires tripled from 2007 to 2017 (representing from 6% in 2007 to 26% in 2017 of total primary $PM_{2.5}$ emissions) with significant interannual variability as ambient $PM_{2.5}$ levels declined (Figure 4B). It has been previously shown that wildfires smoke concentrations were strongly related the frequency and magnitude of wildfires in eastern US.[13–36] In this analysis, industrial and residential wood combustion were included in the industrial sector emissions.

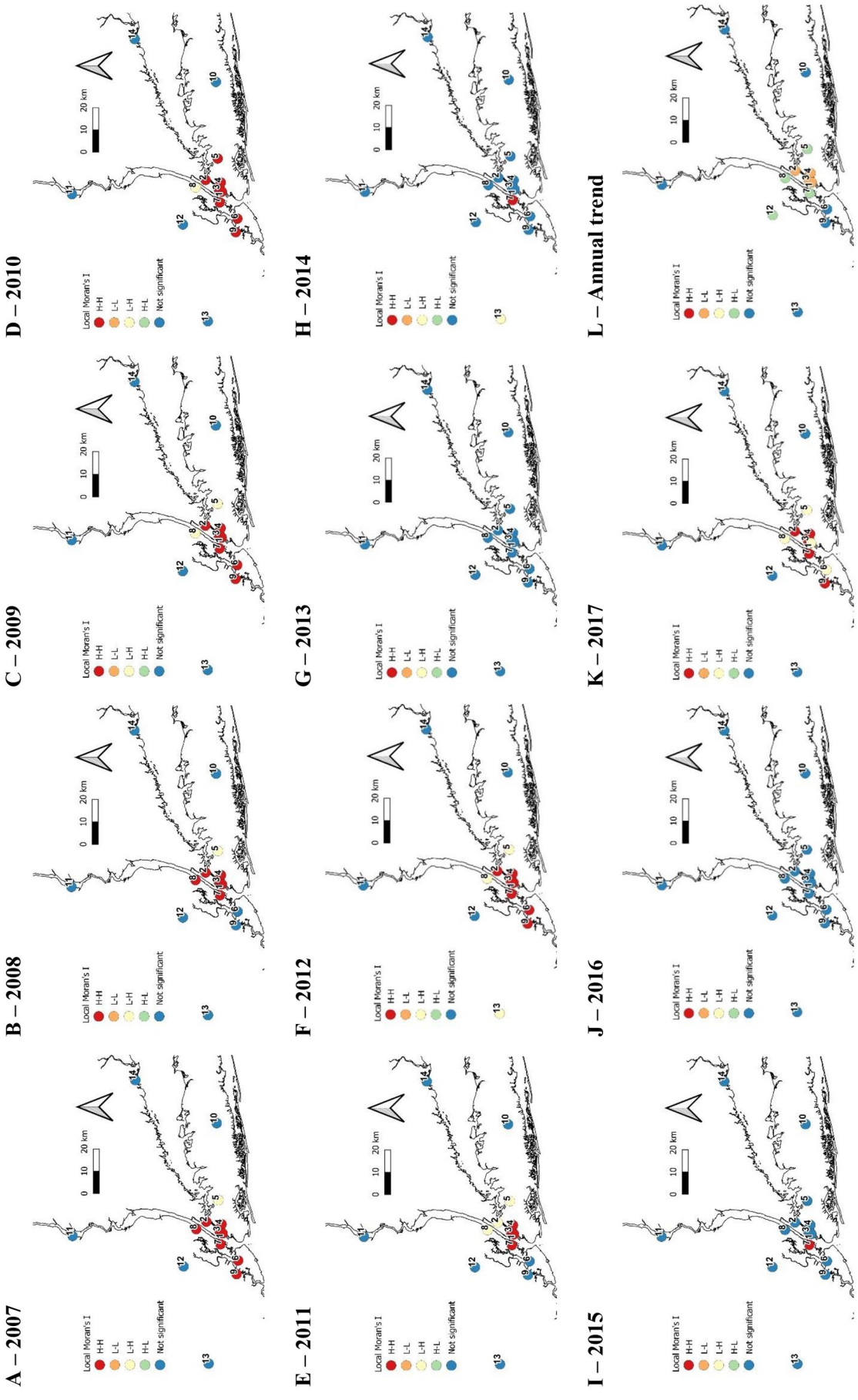

Figure 3. Local spatial autocorrelation of (A-K) $PM_{2.5}$ pollution from 2007 to 2017 and (L) $PM_{2.5}$ annual trends.

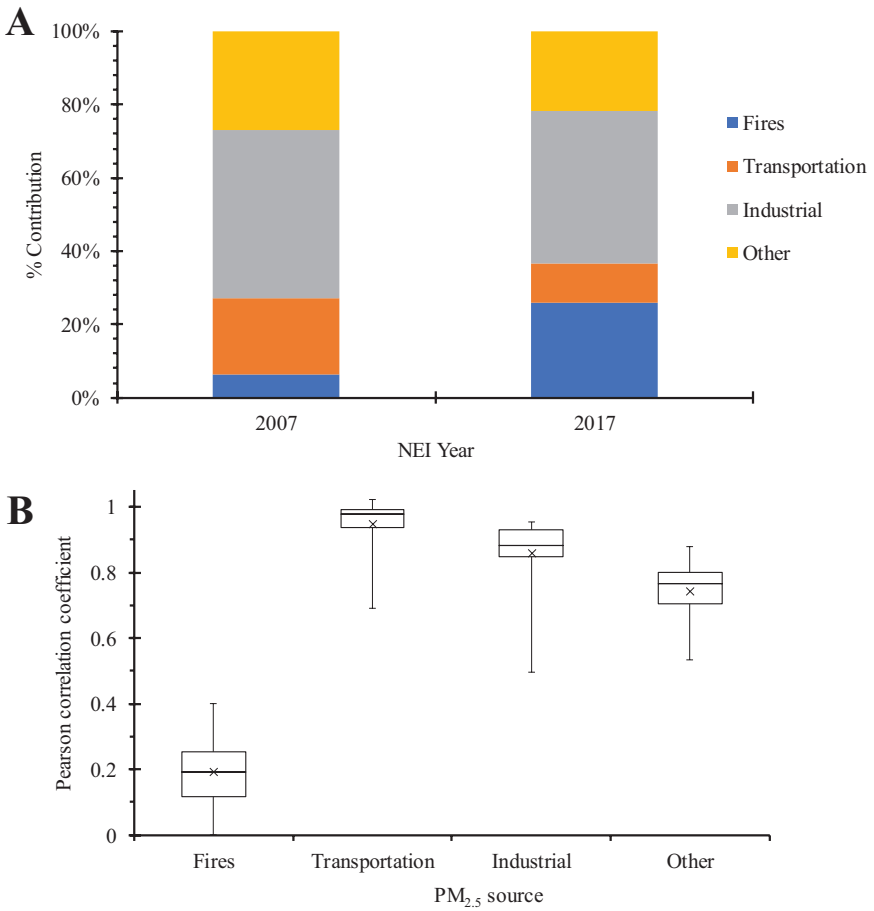

Figure 4. The relative abundance of primary $PM_{2.5}$ emissions in 2007 and 2017 (A) and site-specific Pearson correlation coefficients (B) from fires, industrial sources, transportation, and other sources in NY, NJ, and CT during the 2007 to 2017 period. The horizontal line denotes median values, boxes extend from the 25th to the 75th percentile of sources; × denotes the mean value; vertical extending lines denote minimum and maximum values.

According to the 2014 NEI, 14 000 metric tons of $PM_{2.5}$ (one-third of industrial emissions) were emitted from biomass burning accounting for approximately 93% of residential wood combustion.[37] The New York State Energy Research and Development posited that bioenergy particularly the use of wood as a primary heating source fluctuated between 2002 and 2012.[38] Between 2005 and 2012, the number of homes using wood as the primary heating source in New York State increased by 60% but leveled off by 2012.[37,38]

Considering that secondary inorganic species accounted for 46% to 57% in 2007, 25% and 46% of $PM_{2.5}$ in 2017,[19] the contribution of primary $PM_{2.5}$ emissions on the remaining ambient $PM_{2.5}$ mass declined by 0.8 $\mu g/m^3$ for transportation (from 1.3 $\mu g/m^3$ in 2007 to 0.5 $\mu g/m^3$ in 2017), 0.9 $\mu g/m^3$ (from 2.8 $\mu g/m^3$ in 2007 to 2.0 $\mu g/m^3$ in 2017) for industrial sources and 0.6 $\mu g/m^3$ (from 1.7 $\mu g/m^3$ in 2007 to 1.0 $\mu g/m^3$ in 2017) and increased by 0.8 $\mu g/m^3$ (from 0.4 $\mu g/m^3$ in 2007 to 1.2 $\mu g/m^3$ in 2017) for local fires. The approach infers that $PM_{2.5}$ (other than sulfate and nitrate) concentration are proportionally related to local $PM_{2.5}$ emissions and the relationship did not change over time. There are several limitations in this study. First, other local sources not included in the EPA NEI system may contribute to $PM_{2.5}$ mass. They may include soil dust, sea salt, marine emissions and recreational biomass burning and barbequing. These area sources may account for less than 5% of $PM_{2.5}$ mass and there were not subject to policy controls and regulations. Another aspect of our study, that probably underestimates the contribution of fires on $PM_{2.5}$ mass is the use statewide emissions for all sites. Sites located close to the fires, usually perimetrically to urban along the wildland-urban interface are most likely to experience higher $PM_{2.5}$ levels than downwind locations. Lastly, the intra-annual variability of synoptic scale weather systems may affect the relationship between $PM_{2.5}$ emissions and ambient $PM_{2.5}$ levels. The effect of this may be offset by using annual measurements. It provides a conservative and empirical estimate of the contribution of primary $PM_{2.5}$ emissions. Transported smoke aerosols may account for up to 5 $\mu g/m^3$ in $PM_{2.5}$ in New York State.[36] An increase of 2.2 $\mu g/m^3$ of $PM_{2.5}$ mass at the Pinnacle background site in NY was assigned to biomass burning emissions.[30]

These changes in the relative abundance of primary $PM_{2.5}$ sources emphasize the need to better understand the local and regional drivers of air pollution including the role of climate change. The El Niño-Southern Oscillation (ENSO) is shown to modifying the frequency and intensity of wildfires in the

US.[39,40] It suggests that the chemical content of $PM_{2.5}$ may be transitioning from mostly inorganic species (secondary sulfate and nitrate) to a mixture of carbonaceous (elemental and organic carbon) aerosol. The chemical content of biomass burning smoke may change over time and space. Fresh biomass burning and woodburning smoke contains mostly aromatic species (up to 80%).[41] During transport, smoke may undergo photochemical aging, leading to significant changes in the chemical composition including the formation of carbonyl and carboxyl-compounds and polyaromatics, decreasing aromatic and heavy metals content.[41,42] As a result, toxicological responses and mechanisms including changes in cell metabolic activity and cell death by apoptotic and necrosis pathways may be differentiated.[42] Overall, the abundance of primary and secondary organic aerosol from wildfires and domestic biomass burning on ambient $PM_{2.5}$ mass may be increasing. Because of the coupling with regional atmospheric processes and global climate dynamics, emissions from these sources may be difficult to manage and control.

Conclusion

The spatiotemporal patterns and trends of $PM_{2.5}$ in the NYNJ MSA over the 2007 to 2017 period were examined. Daily $PM_{2.5}$ mass concentrations were retrieved from 14 sites within the US EPA air quality network located in urban and peri-urban locations. $PM_{2.5}$ mass concentrations decreased across all sites, with slightly faster declines for sites located in heavily populated areas. A strong urban-periurban gradient in 2007 gradually declined by 2013. This decline was consistent with national and regional trends and was attributed to reductions of gaseous precursors of particulate sulfate and nitrate from industrial and anthropogenic sources. This is consistent with the trend of sulfate and nitrate measurements in speciated $PM_{2.5}$ and modeled estimates of transported inorganic aerosol in New York City. $PM_{2.5}$ reductions slowed down during the 2013 to 2017 period, accompanied by a feeble spatial gradient. Increasing primary $PM_{2.5}$ emissions from fires during the same period indicated that the contribution of biomass burning on ambient $PM_{2.5}$ may be increasing. This trend may imply changes in the content of fine particles, from ammonium sulfate and ammonium nitrate salts to hazardous carbonaceous aerosol, the composition of which varies by time and location due to continuous photochemical aging during transport from the fire to the receptor site.

Author Contributions

IGK and SS conceived and designed the study. SS analyzed the data and wrote the first draft of the manuscript. GJ and IGK contributed to the writing of the manuscript and given critical comments on the draft manuscript. All authors agree with manuscript results and conclusions, reviewed, and approved the final manuscript.

Supplemental Material

Supplemental material for this article is available online.

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