

**LOCAL VERSUS UPWIND CONTRIBUTIONS TO
PM_{2.5} MASS & ELEMENTAL CONSTITUENT
CONCENTRATIONS IN NEW YORK CITY**

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**NEW YORK STATE
ENERGY RESEARCH AND
DEVELOPMENT AUTHORITY**





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Final Report

Prepared for the
**NEW YORK STATE
ENERGY RESEARCH AND
DEVELOPMENT AUTHORITY**

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PREFACE

The New York State Energy Research and Development Authority is pleased to publish “Local vs. Upwind Contributions to PM_{2.5} Mass and Elemental Constituent Concentrations in New York City.” The report was prepared by George Thurston and Ramona Lall of New York University.

This work was funded by the New York Energy Smart Environmental Monitoring, Evaluation, and Protection (EMEP) Program. This study is one of a broader portfolio of research projects characterizing particulate matter (PM), performing source apportionment on PM datasets, and addressing policy-relevant questions for PM control strategies in New York State.

NOTICE

This report was prepared by Dr. George D. Thurston and Ms. Ramona Lall of the New York University School of Medicine (NYU SOM) in the course of performing work contracted for, and sponsored by, the New York State Energy Research and Development Authority (hereafter "NYSERDA") under NYSERDA Agreement 6084-ERTER-ES-99. The opinions expressed in this report do not necessarily reflect those of NYSERDA or the State of New York, and reference to any specific product, service, process, or method does not constitute an implied or expressed recommendation or endorsement of it. Further, NYSERDA, the State of New York, and the contractor make no warranties or representations, expressed or implied, as to the fitness for particular purpose or merchantability of any product, apparatus, or service, or the usefulness, completeness, or accuracy of any processes, methods, or other information contained, described, disclosed, or referred to in this report. NYSERDA, the State of New York, and the contractor make no representation that the use of any product, apparatus, process, method, or other information will not infringe privately owned rights and will assume no liability for any loss, injury, or damage resulting from, or occurring in connection with, the use of information contained, described, disclosed, or referred to in this report.

ABSTRACT

New York City is presently in violation of the nation's fine particle ($PM_{2.5}$) annual mass standard under the Clean Air Act, and will have to take actions to control the sources contributing to these violations. This report seeks to differentiate the impact of long-range transported aerosols on the air quality of downtown New York City (NYC), so that the roles of local sources can be more clearly evaluated. Previous source apportionment studies have used single sites individually in their analyses to identify and determine local and non-local sources affecting that site. In this study, a rural site located in Sterling Forest, NY, which is near the New York City area but unaffected by local New York City sources, is used as a reference to separate the portion of the aerosol that is transported to a sampling site at Hunter College in Manhattan, NYC, when conducting the source apportionment analysis. In this study, Sterling Forest is confirmed as a background site via elemental comparisons with NYC during regional transport episodes of Asian and Sahara desert sandstorm dusts, as well as by comparisons with a second background Speciation Trends Network site in Chester, NJ, and by examinations of wind back-trajectories on sampling days. In the source apportionment analysis of contributions to $PM_{2.5}$ at the Hunter College site in 2001, we subtracted Sterling Forest impacts from the NYC source apportionment analysis for the daily $PM_{2.5}$ elemental and mass concentration data, in order to separately estimate local versus transported pollution. Six $PM_{2.5}$ source categories are identified for NYC: (1) regionally transported sulfate; (2) trans-continental desert dust; (3) mobile sources (or traffic); (4) residual oil combustion (e.g., from power plants or commercial boilers burning residual fuel); (5) local wind-blown dust; and (6) the pollution impact from the World Trade Center fires (primarily on Sept. 12, 2001). Of these source contributions, the transported sulfates account for nearly half of the total $PM_{2.5}$ mass at the Hunter College site during 2001. During the summer months, nearly two-thirds of the $PM_{2.5}$ mass were attributed to transported (non-local) sources. The highest $PM_{2.5}$ days at Hunter College, NYC, were also those where concentrations of sulfate were the highest. There were differing percentages of the various elements that were transported versus produced locally. Virtually all of the elemental carbon at the Hunter College site was of local origin, while nearly all of the sulfate mass was transported into the city. The two major local sources identified were traffic-related pollution and residual oil combustion. While the former was found to impact air quality on a year-round basis, with higher levels in the summer, the latter was found to have its highest impacts in winter and a negligible impact in summer. Residual oil combustion was the largest contributor among all sources during the winter months. Our results indicate that transported pollution plays a major role in NYC's fine-PM pollution. Any efforts to meet the ambient $PM_{2.5}$ air quality standards in NYC will require that upwind sources of sulfates outside of the city be controlled. Although local combustion and traffic sources in NYC individually contribute less than transport on an annual average basis, controlling these local sources would also greatly benefit air quality and health in NYC.

Keywords: Source apportionment, $PM_{2.5}$, New York City, PM speciation data, reference site, up-wind vs. local sources, long-range transported sulfate aerosols, traffic, residual oil, trans-continental desert dust.

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EXECUTIVE SUMMARY

Numerous epidemiological studies indicate associations between particulate matter (PM) pollution and both human morbidity and mortality (e.g., USEPA, 2003a). However, PM mass concentrations are chemically non-specific, and the PM-health associations observed in these studies could indicate toxicity of certain types of particles in specific size ranges, rather than toxicity related to mass itself. The growing consensus among the scientific community is that both particle size and PM chemical constituents, considered together, are important in understanding the toxicity of PM. Fine particles less than 2.5 micrometers (μm) in aerodynamic diameter (i.e., $\text{PM}_{2.5}$) and their precursors are emitted from various sources and have differing characteristic compositions, including varying levels of elemental carbon, metals, sulfates, and organics. It is critical to define the sources and types of PM that most negatively affect human health so that we can select and apply the most appropriate control strategies. Previous studies have established that transported particulate matter (PM) aerosols have been a major contributor to air pollution in New York City (NYC) and the northeastern United States. However, quantifying exactly how much PM is local in origin and how much is transported from outside NYC has been challenging. This report outlines research results from a NYSERDA-funded research project regarding $\text{PM}_{2.5}$ concentrations in Manhattan (NYC) and their local and up-wind sources. This study uses two monitoring sites in the NYC metropolitan area funded by NYSERDA to collect $\text{PM}_{2.5}$ filter samples on a daily basis for subsequent chemical speciation analysis.

The hypothesis we posited at the start of this research project was that “local power plant and industry sources will have a much smaller impact than transported aerosols on the New York City $\text{PM}_{2.5}$ concentrations.” Such a distinction between local and non-local sources, and assessment of the total impact of transported pollution into the city, could provide more insight to the air pollution problems in NYC, and could thereby have important policy ramifications on the control strategies employed to meet the new National Ambient Air Quality Standards (NAAQS) for $\text{PM}_{2.5}$ in New York State under the federal Clean Air Act.

In order to estimate the roles of local versus transported pollution sources for NYC, we applied source apportionment techniques, including positive matrix factorization (PMF), to quantify the mass contributions from different $\text{PM}_{2.5}$ pollution sources. In the receptor-oriented

be relatively unaffected by local or NYC pollution. This assumption was tested by comparing the elemental concentrations in the two locales. For instance, while sulfur concentrations at both the NYC and Sterling Forest sites are very similar throughout the year, large differences in concentrations of elemental carbon are found between the sites (Table E1). This evidence suggests that sulfate aerosols affect both sites similarly, while the aerosols locally produced in NYC do not affect Sterling Forest. In addition, we determined that Sterling Forest was a good background site for the study of transport aerosol episodes, such as during the trans-continental transport of desert dust from the Gobi and Sahara deserts in April 2001 (USEPA, 2003b). Based on these observations, the study determined that Sterling Forest was a suitable background site to the Hunter College site.

Table E1: Mean annual and seasonal concentrations (Standard Deviations) of EC, OC, S, and PM_{2.5} measured at the two NYU Sampling Sites during 2001 (in $\mu\text{g}/\text{m}^3$)

	2001	Sterling Forest	Hunter College
Elemental Carbon	Annual	0.2 (0.2)	1.2 (0.6)
	Winter	0.2 (0.1)	1.3 (0.7)
	Summer	0.2 (0.2)	1.3 (0.6)
Organic Carbon	Annual	1.7 (1.0)	3.6 (1.3)
	Winter	1.3 (0.5)	2.7 (0.9)
	Summer	2.9 (1.3)	4.4 (1.3)
Sulfur	Annual	1.5 (1.4)	1.6 (1.3)
	Winter	1.1 (0.6)	1.2 (0.5)
	Summer	2.6 (2.2)	2.5 (1.9)
PM_{2.5}	Annual	11.4 (8.5)	17.3 (9.6)
	Winter	8.8 (5.4)	17.3 (9.0)
	Summer	18.3 (11.0)	21.4 (11.9)

Winter = January – March; Summer = June – August.

Air pollution at urban sites is a complex mixture of aerosols produced locally as well as those transported from great distances. This often complicates the discernment of sources, especially when much of the mass is transported regionally. In this study, a new approach is used, wherein data from an additional background site (Sterling Forest) are incorporated into the NYC source apportionment PMF model in order to quantitatively estimate the roles of transported versus local aerosols. This technique assumes that all of the sulfur measured at

Sterling Forest is transported into the region, and that the transported sulfur will similarly affect the Hunter College site. Furthermore, the remainder of the NYC sulfur (over and above that seen in Sterling Forest) is assumed to be of local origin and therefore contributed by the local PM_{2.5} sources in NYC. This conclusion is based upon the validated assumption that very little PM pollution is transported from NYC to Sterling Forest, and that the transported sulfate pollution that affects NYC also impacts the Sterling Forest site. Thus, in addition to the 20 elements derived from X-ray fluorescence (XRF) of the samples and organic and elemental carbon measurements from the Hunter College site, two sulfur variables were included as a part of this apportionment analysis: both a “transported sulfur” variable and a “local sulfur” variable, where:

$$\begin{aligned} S_{\text{TRANSPORTED}} &= S_{\text{SF}} \\ S_{\text{LOCAL}} &= S_{\text{NYC}} - S_{\text{SF}} \end{aligned}$$

Six PM_{2.5} source categories are thereby identified for NYC: (1) regionally transported sulfates; (2) trans-continental desert dust; (3) mobile sources (or “traffic”); (4) residual oil combustion (e.g., from power plants or commercial boilers burning residual fuel); (5) local wind-blown dust; and (6) the pollution impact from the World Trade Center (WTC) fires (primarily on Sept. 12, 2001) (see Table E2 for PM_{2.5} contributions from each source). This approach identified similar sources as were found in a conventional source apportionment of the NYC data alone (i.e., without subtracting out the Sterling Forest sulfur concentrations). By incorporating the concentrations from a background site in the analysis, we were able to more clearly identify the observed sulfate-associated PM_{2.5} mass from upwind (non-NYC) sources. Past studies that have conducted source apportionment of a single site (without incorporating concentrations from a background site) have similarly identified a “secondary sulfate” source factor (e.g., Thurston and Lioy, 1987). However, such studies were unable to definitively attribute such a source factor entirely to transport. By incorporating sulfur data from a background site, we are able to identify and quantify the transported sulfate PM_{2.5} component with greater confidence, since we do not assume that all of the sulfate-related impacts are caused by transport alone.

Table E2: The annual and seasonal mass contributions (in $\mu\text{g}/\text{m}^3$) for 2001 (95% CI of contribution estimates) for the six sources identified for NYC using the “two sulfur variables” PMF model

2001 PM _{2.5} MASS CONTRIBUTIONS	TRANSP. SULFATES	TRAFFIC	RESIDUAL OIL	TRANSP. DESERT DUST	Fe-Mn or "LOCAL" DUST	WTC
ANNUAL ($\mu\text{g}/\text{m}^3$)	7.9 (7.4 - 8.4)	6.7 (5.9 - 7.5)	3.4 (3.0 - 3.9)	1.1 (0.8 - 1.4)	0.4 (0.1 - 0.6)	0.4 (0.4 - 0.5)
WINTER (JAN-MAR '01)	6.4 (5.9 - 6.8)	4.8 (4.2 - 5.3)	7.7 (6.7 - 8.7)	0.5 (0.3 - 0.6)	0.2 (0.1 - 0.3)	0.3 (0.3 - 0.4)
SPRING (APR-JUN '01)	8.7 (8.1 - 9.2)	6.4 (5.7 - 7.1)	2.4 (2.1 - 2.7)	2.1 (1.5 - 2.7)	0.2 (0.1 - 0.3)	0.2 (0.2 - 0.2)
SUMMER (JUL-SEP '01)	10.5 (9.8 - 11.2)	8.3 (7.4 - 9.2)	0.9 (0.8 - 1.1)	0.8 (0.6 - 1.1)	0.4 (0.1 - 0.7)	0.8 (0.7 - 1.0)
FALL (OCT-DEC '01)	5.3 (5.0 - 5.7)	6.8 (6.0 - 7.6)	3.7 (3.3 - 4.2)	0.9 (0.6 - 1.1)	0.8 (0.3 - 1.3)	0.4 (0.3 - 0.5)

This work, like previous studies, also found that PM_{2.5} and sulfate aerosols have a major impact on NYC’s air quality. The 2001 annual estimate of PM_{2.5} mass concentrations ($17.3 \mu\text{g}/\text{m}^3$) for NYC was above the federal annual standard ($15.0 \mu\text{g}/\text{m}^3$). As summarized in Table E3, when comparing PM_{2.5} mass and its components between the two sites on an annual basis, about 45% of the total PM_{2.5} measured in NYC is a result of sulfate aerosols being transported into the city; during the summer months, the fraction due to transport can be as much as two-thirds. For sulfates, over 90% of sulfates affecting the Hunter College site can be attributed to transport. By contrast, 95% of elemental carbon at the Hunter College site is determined to be of local origin. Organic carbon, in contrast, is determined to be more equally attributed to local (~70%) and non-local (~30%) pollution sources. The major transported aerosol contributions to NYC of total PM_{2.5}, sulfates, and organic carbon suggest that if the NYC metropolitan area is to reliably meet the PM_{2.5} standards, then one important course of action will be to specifically target regional PM_{2.5} reductions. Along with alleviating PM_{2.5} concentrations in the northeastern United States and Canada, these measures would also improve visibility and reduce acid deposition in sensitive ecological areas in the region.

Table E3: Estimated fractions of total PM_{2.5} mass and elemental/carbon concentrations contributed by transported aerosols in NYC

	ANNUAL (2001)	WINTER (Jan-Mar '01)	SUMMER (Jun-Aug '01)
PM _{2.5}	0.44	0.33	0.56
S	0.80	0.79	0.85
OC	0.31	0.26	0.67
EC	0.12	0.08	0.15

This research finds that the highest PM_{2.5} days in NYC occur when contributions from transported sulfate aerosols are also the largest. In fact, on some days between June and August, concentrations of transported sulfate aerosols alone were over 35 µg/m³ (i.e., exceeding the newly revised 24-hour PM_{2.5} mass standard). Aside from the transported sulfate aerosols, two other primarily local sources were found to contribute significantly to NYC's air pollution: traffic and residual oil. However, the only major fine particle PM_{2.5} mass contribution to this mid-Manhattan site from WTC-related fires or rescue/clean-up activities at Ground Zero identified in this research was the direct plume of the WTC fires measured on September 12, 2001 at the Hunter College site. The relatively cleaner air quality days (i.e., when the 10th percentile of PM_{2.5} mass concentrations were less than 7 µg/m³), had greater contributions from traffic and residual sources, compared to transported sulfates. Therefore, the contributions of these local sources cannot be overlooked when seeking to control ambient pollution exposures in the City of New York. In addition, this study has found that utilizing additional data from a nearby reference site (largely unaffected by local pollution sources) as an indicator of transported pollution is a helpful approach to more clearly identify local versus upwind source contributions to PM_{2.5}.

INTRODUCTION

Epidemiological studies indicate associations between particulate matter (PM) air pollution and human morbidity and mortality (e.g., USEPA 2003a). Until recently, most research studies focused on particle size, especially particles in the “fine” fraction. Currently, the PM standards are based on the mass concentration of two PM size fractions, PM₁₀ (particulate matter less than 10 micrometers in diameter) and PM_{2.5} (particulate matter less than 2.5 micrometers in diameter). Studies have also found health effects in cities that meet the PM_{2.5} standard (Vedal, 2003). The PM_{2.5} standard inherently assumes that all particles are equally harmful and that there exists a threshold above which human health will be affected. However, particles and their precursors are emitted from various sources and therefore have differing characteristic compositions, including varying levels of elemental carbon, metals, sulfates, and organics. The Harvard Six-City study found that combustion-related particles (e.g., particles from vehicular exhaust and electric utilities) are generally more harmful than other particles (e.g., soil), thereby suggesting that some particles are more injurious to human health than others (Laden et al., 2000).

Therefore, chemically non-specific PM mass concentrations alone cannot provide a complete indication of PM toxicity. Instead, a combination of size and chemical constituents of PM could prove to be more useful in explaining the PM-health associations observed previously. In 2000, the United States Environmental Protection Agency (USEPA) Speciation Trend Network was initiated to monitor PM_{2.5} and its elemental constituents at over 200 sites across the United States (USEPA, 1999). Around the same time, NYSERDA funded a study with New York University (NYU) to collect daily PM_{2.5} filter samples at two sites in the New York City (NYC) Metropolitan Area (Hunter College and Sterling Forest) for chemical analysis. The purpose of this study was to characterize and quantify PM_{2.5} and its chemical constituents from local and upwind non-local sources to Manhattan, NYC. This report outlines results from the study. Future studies that use such speciation techniques could potentially identify sources and types of PM most affecting air quality and health, which could potentially lead to more appropriate air pollution control policies and air quality standards.

The effect of sulfate aerosols transported into NYC and much of the northeastern United States on the air quality of this region has been well studied and documented (Lippmann, 1979; Liou, 1980). For example, the New York Summer Aerosol Study, conducted almost 30 years

ago, concluded that, on certain summer days, most of the sulfate and a significant fraction of total suspended particulate matter (TSP) could be attributed to long-range transport from sources in the midwestern United States (Lioy et al., 1980). As a result of this extensive multi-site study, an estimated 73% of measured sulfate at that time was attributed to transport into the NYC region.

Although regional air pollution emissions and concentrations have changed over the past three decades, and are generally lower today, long-range transported aerosols still contribute a significant portion of particulate air pollution in NYC. Data from the aforementioned Speciation Trend Network (STN) have been used to try to assess the role of transport in recent years. A study using the STN data for 2001-2002 attributed approximately half the sulfate measured at the Queens (NY) STN site to transport (Dutkiewicz et al., 2003). This estimate was based on data from a single site and considered variations in sulfate concentrations as a function of the wind direction. Sulfur levels for each of the wind direction sectors were computed, and the authors assumed that the sectors with the lowest sulfur levels were unaffected by transport and were instead indicative of the local contributions to total sulfate. The authors subtracted the local sulfur concentration from the sulfur levels from each of the various wind quadrants to estimate the overall sulfate fraction from transported aerosols. However, it is certainly possible that the local sulfur identified by this process also included some transported sulfur. This could easily result in an overestimation of local sulfur and an underestimation of the transported sulfur. Therefore, these estimates are probably best viewed as minimum estimates of transported pollution impacts in NYC.

The STN data for the NYC metropolitan area have also been used in other source apportionment studies (Ito, 2004, and Qin et al., 2006). It should be noted that the STN data are collected every three days, and, although both studies used multiple sites in their analysis, none of those sites were located in Manhattan. Both studies typically analyzed for sources at each particular site, and thereafter compared sources resolved and their contributions at the sites. Qin et al. included a background site (located in Chester, NJ) in their analysis. By differentiating between the background site at Chester and the STN data from New York City, they estimated that 69-82% of $PM_{2.5}$ is transported to NYC from long-distance sources.

This study advances this differencing approach by directly incorporating the aerosol information from a simultaneously monitored rural background site (unaffected by local NYC

sources) into the Hunter College site source apportionment factor analysis, in order to quantitatively differentiate and quantify the transported and local fine particle pollution at the Hunter College site.

In order to determine local versus transported pollution, we apply positive matrix factorization (PMF) source apportionment analysis to quantify the mass contributions from different sources of $PM_{2.5}$. In the receptor-oriented source apportionment modeling, concentrations of various species (i.e., trace elements and carbon measurements) at the ambient sampling site (the receptor) are analyzed in order to: (1) identify the major particle sources; (2) determine their appropriate source profiles; and (3) obtain their respective contributions to the mass (i.e., provide a total mass source apportionment). In this approach, PMF produces factors that, based on their individual elemental characteristics and known characteristics of $PM_{2.5}$ sources, are then identifiable as distinct $PM_{2.5}$ source categories. However, individual sources (e.g., individual power plants) are not identifiable using this method. These apportionment methods have been well documented in the literature over the past few decades (e.g., Cooper and Watson, 1980; Hopke, 1985; Henry, 1991).

METHODS: DATA COLLECTION & CALIBRATION PROCEDURES

As part of a NYSERDA-funded project, NYU set up two PM monitoring sites in the NYC metropolitan area and began sampling $PM_{2.5}$ and its chemical constituents on a daily basis in January 2001. One site was situated in Manhattan, at Hunter College on 26th Street and 1st Avenue, and the other site was located at the Nelson Institute of Environmental Medicine, Sterling Forest, in Tuxedo, NY, approximately 45 miles northwest of NYC (see Figure 1). Sterling Forest is a rural site surrounded by thousands of acres of largely undeveloped woodland within New York State's Sterling Forest State Park, which has an area approximately the size of Manhattan (see Figure 2). As indicated by the NYC wind rose for Central Park (National Climatic Data Center, 2007) shown in Figure 3, the Sterling Forest site is seldom affected by New York City pollution (i.e., only during infrequent winds from the south-southeast). Thus, the purpose of locating a site in Sterling Forest was to provide a background reference for the Hunter College site that has none of the NYC local source impacts but is similarly affected by

Figure 1: Location of the NYU and EPA STN monitors

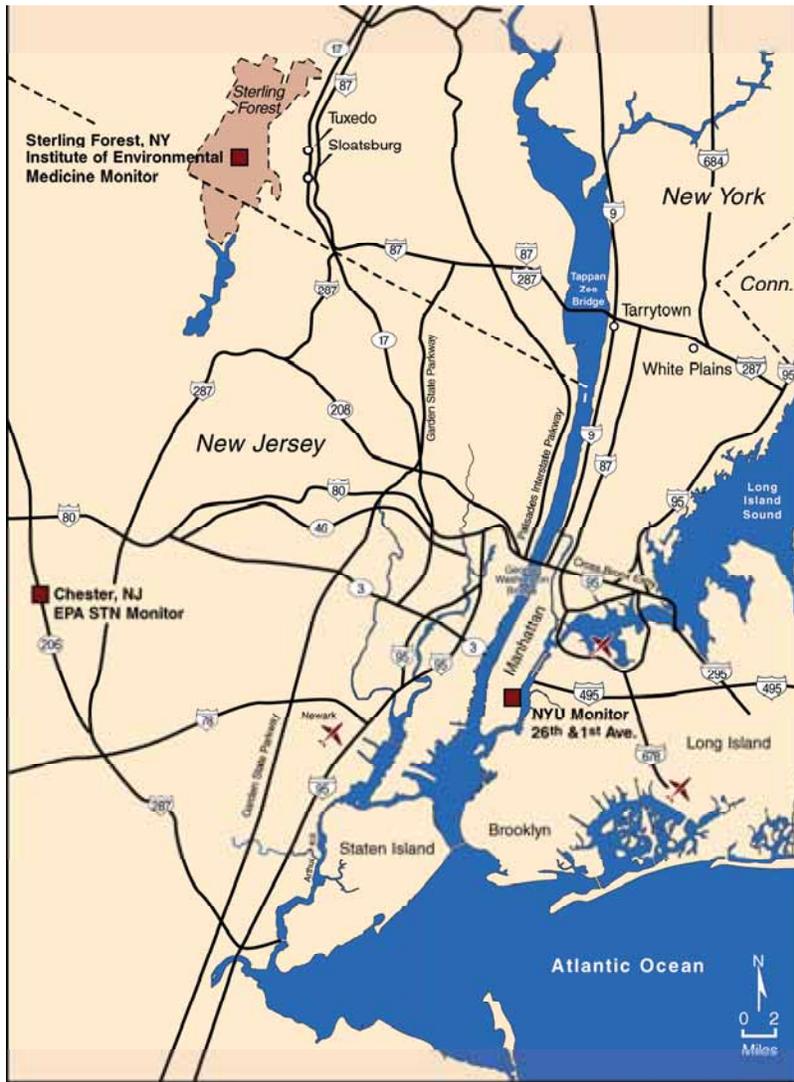


Figure 2: Monitoring site at Sterling Forest, (Tuxedo, NY) looking west-southwest

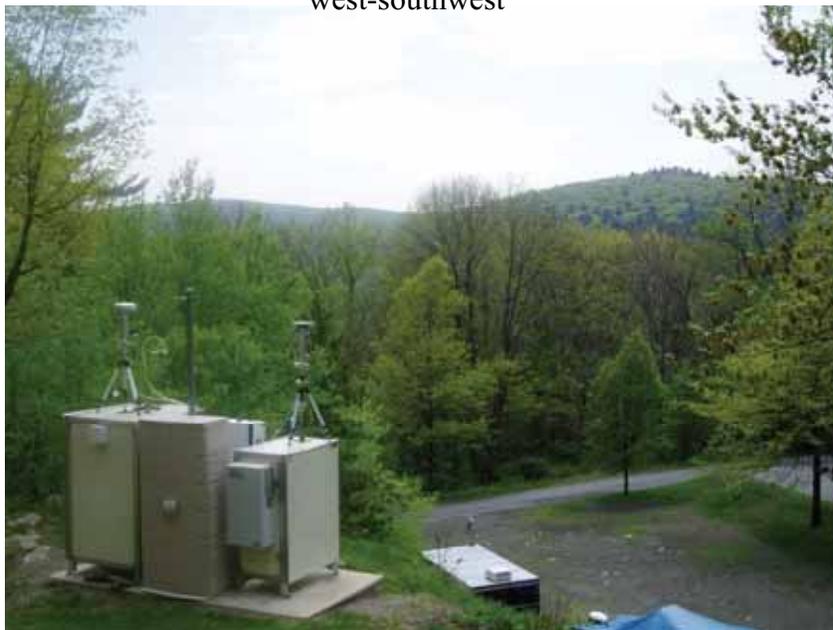
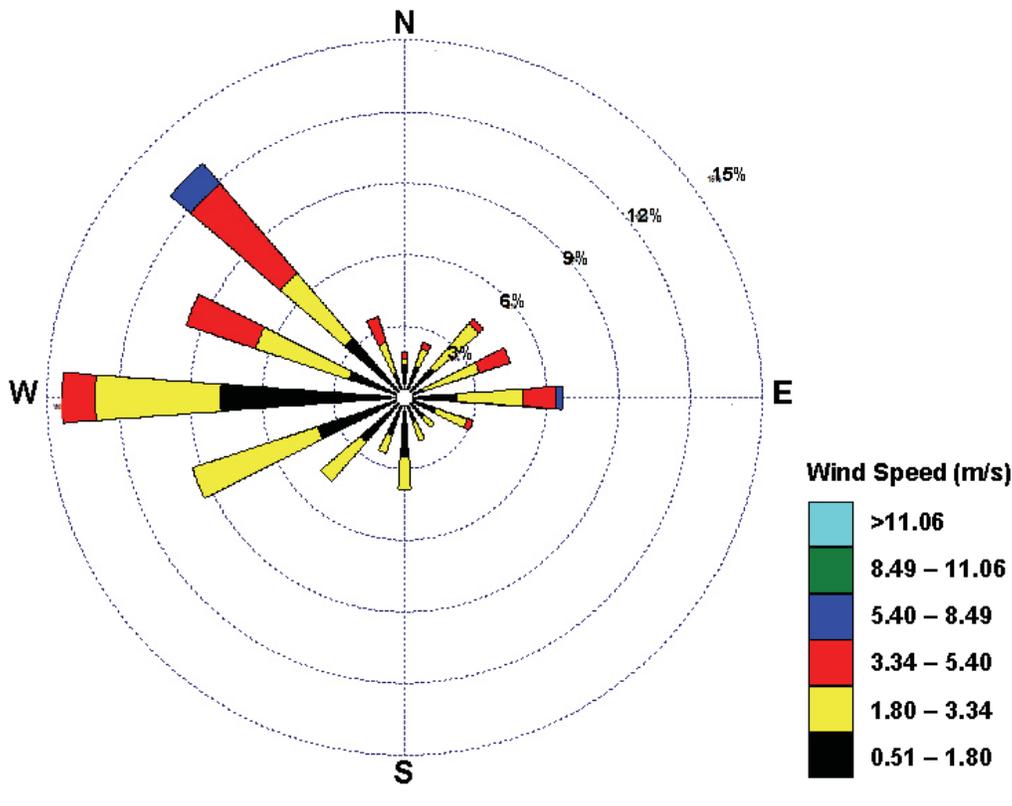


Figure 3: Wind rose showing the frequency of daily mean wind speed and direction of winds (i.e., direction blowing from) at Central Park, NYC in 2001



transported pollution (largely carried into the NYC metropolitan area by winds from the west and southwest). This assumption has been tested in this study using back-trajectory analysis and comparisons between sites (both background and urban).

At both sites, 24-hour $PM_{2.5}$ cumulative filter samples were collected, and continuous real-time $PM_{2.5}$ mass concentration data were collected every half-hour. Elemental carbon (EC) and organic carbon (OC) levels were also measured every 3 hours. Neither nitrates nor pollutant gases were monitored at these sites, and therefore data for these pollutants are not available for consideration in this work. The following is a brief description of the instruments used at both monitoring sites and the quality control procedures followed.

Particulate mass and elemental composition sampling

Twenty-four-hour cumulative $PM_{2.5}$ filter samples were collected using an Automated Cartridge Collector Unit (ACCU) sampler at each of the sites, while semi-continuous fine particle concentrations were recorded via a Tapered Elemental Oscillating Microbalance (TEOM) sampler (Rupprecht & Patashnick). In this work, the TEOM data are not used directly in the analysis; rather, these additional PM data (available for every half-hour) are consulted

during quality assurance when investigating particular days (e.g., Sept. 12, 2001) or apparent outliers in the filter data. The ACCU and the TEOM were designed by their manufacturer to share a common PM_{2.5} inlet, and the inlet size cut-point is equivalent to the PM_{2.5} reference method. The pump was operated at 16.7 LPM, with the TEOM sampling air at 3 LPM, and the remainder air flow (13.7 LPM) being bypassed to the ACCU sampler filters. The ACCU sampler contains eight airflow channels to which cartridges holding 37 mm Teflon filters were fitted. The sampler was programmed to sequentially draw ambient air through each channel for 24 hours per sample (midnight to midnight). Each site was visited by an NYU technician on a weekly basis. Prior to the site visit, filters were acclimated and then pre-weighed using a microbalance (Model MT5, Mettler-Toledo Inc., Highstown, NJ). The NYU filter weigh room follows the EPA's filter weighing protocol and maintains a temperature of 21.5°C and relative humidity of 40%. Each week, filters were changed in the cartridges; the filter samples were replaced with a new set of filters (carrying over the filter sampling on the day of collection), and the machine was then programmed to collect 24-hour samples (midnight to midnight) each day for the week ahead. The total volume of air sampled and the elapsed time of sample collection for the previous week were recorded from the ACCU sampler each week. The filter samples collected each week from the site were returned to the lab, where they were post-weighed to determine daily fine particle mass concentration.

The 24-hour filter samples were subsequently analyzed for 34 trace element concentrations using energy dispersive X-ray fluorescence (ED-XRF) techniques (Model EX-6600 –AF, Jordan Valley; Spectral Software XRF2000v3.1 by USEPA and ManTech Environmental Technology, Inc.). Using the USEPA program Spectral Software XRF 2000v3.1 (and ManTech Environmental Technology, Inc), the elemental concentrations and their associated uncertainties were computed. The program includes the error propagation of calibration uncertainty, long-term system stability, uncertainty in least squares fit, uncertainty in attenuation correction, uncertainty in interference correction, uncertainty in flow rate, and uncertainty in sample deposit area. These elemental concentrations and their uncertainties computed by the program were used as a basis for this analysis.

Ambient elemental and organic carbon sampling

R&P ambient carbonaceous particulate monitors (Series 5400), which use a “thermal-CO₂” method, were set up to operate on 3-hour cycles in order to determine EC and OC concentrations. The reference temperatures for OC and EC volatilization were set at 340°C and 750°C, respectively, in accordance with the usual practices of the State of New York in their EC/OC sampling. The instrument was set up to run automatic calibration checks on a weekly basis, and data (along with calibration checks) were downloaded monthly.

METHODS: DATA QUALITY ASSURANCE & CONTROL

The study used the following data quality assurance and control protocol. After filters were weighed and analyzed for their particulate matter chemical constituents, the daily PM data were then aggregated from the various instruments’ results. The data were first screened for days with sampling anomalies, such as those from power failure or targeted flow rates not being achieved. Of the 34 elements analyzed by the XRF, 22 elements were selected for inclusion in the source apportionment analyses. These elements were chosen based on: (1) filter sample concentrations for the element relative to the measurement on the blank filters, as well as compared to the respective levels of detection by the NYU XRF; (2) Signal-to-noise ratios (i.e., elements with larger ratios, typically greater than 2, were identified as “strong” variables and included in further analyses); and (3) extensive exploratory analysis of data, including the investigation of individual time-series plots of trace elements and carbon data. Comparisons of various element concentrations and trends between the NYU data and the USEPA STN datasets were also conducted for quality assurance, providing an additional check against an external database.

Six blank samples were typically run through the XRF with every batch of samples, and these measurements from the blank filters were generally checked after the XRF analysis of the batch was complete. We also compared the average elemental concentrations reported for blank filters between the different batches, to check for XRF detector issues or instrumental drift. Of the 22 elements chosen, only selenium showed some drift. Three batches, towards the latter half of all the analysis, were found to have higher averages of selenium on the blank samples. Rather than lose this important element, the selenium filter concentrations were adjusted by subtracting the blank selenium value from the filter values for these particular batches.

The daily EC and OC concentrations were averaged using data from every 3-hour cycle. Days with less than six out of eight complete 3-hour cycles were removed from the dataset. A standardized 5% of the concentration was used as the uncertainty associated with the measurement. For three weeks in early September 2001, the R&P 5400 monitor (located at the Hunter College site) did not attain the desired temperatures during its analysis cycle, and elemental and organic carbon concentrations are not available for this period, including for September 11th. With these data missing, the trace element concentrations for the same period could not be used by the PMF model. Therefore, the missing carbon data were substituted with an average carbon estimate using data from the month before and after this period. And the uncertainty associated with these substituted values was increased (20% of the estimates), due to their diminished accuracy versus directly measured values.

There are limited elemental data for 2001 from the USEPA STN site at Chester, NJ (with only approximately 50 observations for the entire year). Therefore, these data are used in this work only to test the assumption that Sterling Forest is a suitable background site. USEPA gaseous pollutant data have also been used in this work, to aid in the confirmation of the sources identified. Wind direction data and the on-line HYSPLIT Model from the National Oceanic and Atmospheric Administration (NOAA) were used for supporting meteorological data analysis.

METHODS: DATA ANALYSIS USING SOURCE APPORTIONMENT TECHNIQUES

In this study, the source apportionment of the New York City metropolitan region's pollution was conducted using positive matrix factorization (PMF), a technique based on factor analysis developed by Paatero (1997). This form of factor-based receptor modeling assumes that x_i (i.e., the measured chemical species detected on a given sample) is from p independent pollution sources such that:

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij}$$

where x_{ij} is the j^{th} species concentration measured in the i^{th} sample, g_{ik} is the mass contribution from the k^{th} source on the i^{th} sample, f_{kj} is the j^{th} species mass fraction from the k^{th} source, and e_{ij} is the error term.

PMF uses a weighted least-squares fit, where weights are based on the uncertainties of the elemental concentration measurements (i.e., u_{ij} is the uncertainty estimate in the j^{th} element

measured on the i^{th} sample). PMF provides a solution that minimizes an object function $Q(E)$ based on the uncertainties for each observation and this is defined as:

$$Q(E) = \sum \sum [x_{ij} - \sum g_{ik} f_{kj}]^2 / u_{ij}$$

Unlike other source apportionment models, this model allows for inclusion of measurement uncertainties and thereby provides information regarding the confidence in the concentration measurements into the model fit, which is one of the major advantages of this method versus other factor-based methods. Furthermore, by including non-negative constraints in the model (i.e., that mass contributions or source profile composition fractions cannot be less than 0), only physically plausible solutions are allowed. Numerous source apportionment studies in recent years have successfully employed PMF (e.g., Paatero, 1997; Song, 2001; Ito, 2004; Kim, 2004), and more details on this technique are available in these published papers.

In this study, a different approach to conducting the PMF source apportionment is investigated, one that incorporates background Sterling Forest data into the NYC PMF source apportionment model. In this technique, we assume that all of the sulfur measured at Sterling Forest is transported into the region and that the Hunter College site is similarly affected by this transported pollution. The remainder of the sulfur at the Hunter College site is assumed to be of local origin, and this amount is then apportioned among the local sources using PMF. Therefore, to further aid in the differentiation of transport versus local sources in the source apportionment analysis, we have included a Sterling Forest sulfur variable in the NYC PMF analysis, along with the elemental data from the Hunter College site, and have also computed an estimate of the local sulfur concentration in NYC by differentiating between the two sites. Thus, in addition to the usual 20 elements and the two carbon variables (i.e., OC and EC) for the Hunter College site, two sulfur variables were included as part of this analysis: a “transported sulfur” variable and a “local sulfur” variable, such that, for this Case 1:

$$\begin{aligned} S_{\text{TRANSPORTED}} &= S_{\text{SF}} \\ S_{\text{LOCAL}} &= S_{\text{NYC}} - S_{\text{SF}} \end{aligned}$$

In general, the difference between the Hunter College and Sterling Forest sites (i.e., local sulfur) was found to be slight (mean difference = 85 ng/m³, compared to mean S at the Hunter

College site of 1583 ng/m³ and Sterling Forest mean S of 1512 ng/m³). Furthermore, up to one-third of the observations indicated that sulfur concentrations were higher at the Sterling Forest site compared to the Hunter College site, resulting in negative S_{LOCAL} values. The largest negative differences between the Hunter College and Sterling Forest sites were found to occur during summertime sulfate episodes. These negative contributions are within the measurement error and could be interpreted as being fluctuations around zero or minimal local sulfur contribution. Instead of replacing these negative values with zero, and thereby skewing the local contributions, the differences were included as were in the model. The higher uncertainties (computations shown below) associated with the “local sulfur” ensure that the negative sulfur observations do not influence the model.

Estimates of the elemental uncertainties are required for the application of the PMF model to these data. The uncertainties of elemental concentrations used in the PMF analyses were those reported by the XRF analysis (as described earlier). The Sterling Forest uncertainties were used for the variables classified as “transported sulfur.” For the “local sulfur” variables (calculated by subtracting the Sterling Forest daily concentrations from the Hunter College concentrations on the same day), uncertainties were larger and were propagated from the individual sites’ errors as,

$$U_{\text{LOCAL}} = \sqrt{(U_{\text{SF}}^2 + U_{\text{NYC}}^2)}.$$

To assure the quality of this alternate approach versus the conventional PMF technique, we conducted a PMF source apportionment analysis on the Hunter College site data alone (without subtracting background sulfur concentrations from the Sterling Forest site). We compared the results between the two models to ensure consistency in results (number and types of sources). An additional PMF analysis that extended our approach (by using the background and local differences for all the elements considered in the analysis) was also conducted. Results from this study have been published in the journal *Atmospheric Environment*, where further details are presented (Lall, 2006).

RESULTS

Both the monitoring stations at the Sterling Forest and Hunter College sites began operation in January 2001. This report contains results from data collected and analyzed in 2001.

In this section, brief descriptions of the inter-site comparisons of different PM_{2.5} measurements and results from the source apportionment analysis are provided.

Exploratory analysis of site-to-site variations in PM_{2.5} mass and elemental concentrations

Before attempting the advanced PMF statistical analysis to discriminate the local versus transported aerosol, the two sites' mass and elemental data were compared for indications of the similarities and differences between the two study sites, as well as with other sites in the NYC metropolitan area. The numbers of observations available for the Hunter College and Sterling Forest sites were 331 and 343, respectively. Figure 4 displays the frequency distributions of the PM_{2.5} mass, sulfur, EC, and OC concentrations at the Sterling Forest site in Tuxedo, NY, and the Hunter College site. As shown in Table 1, the site at Hunter College (annual filter mass

Figure 4: Frequency distribution (% of days) of PM_{2.5}, sulfur, elemental carbon and organic carbon concentrations at Sterling Forest and Hunter College for 2001

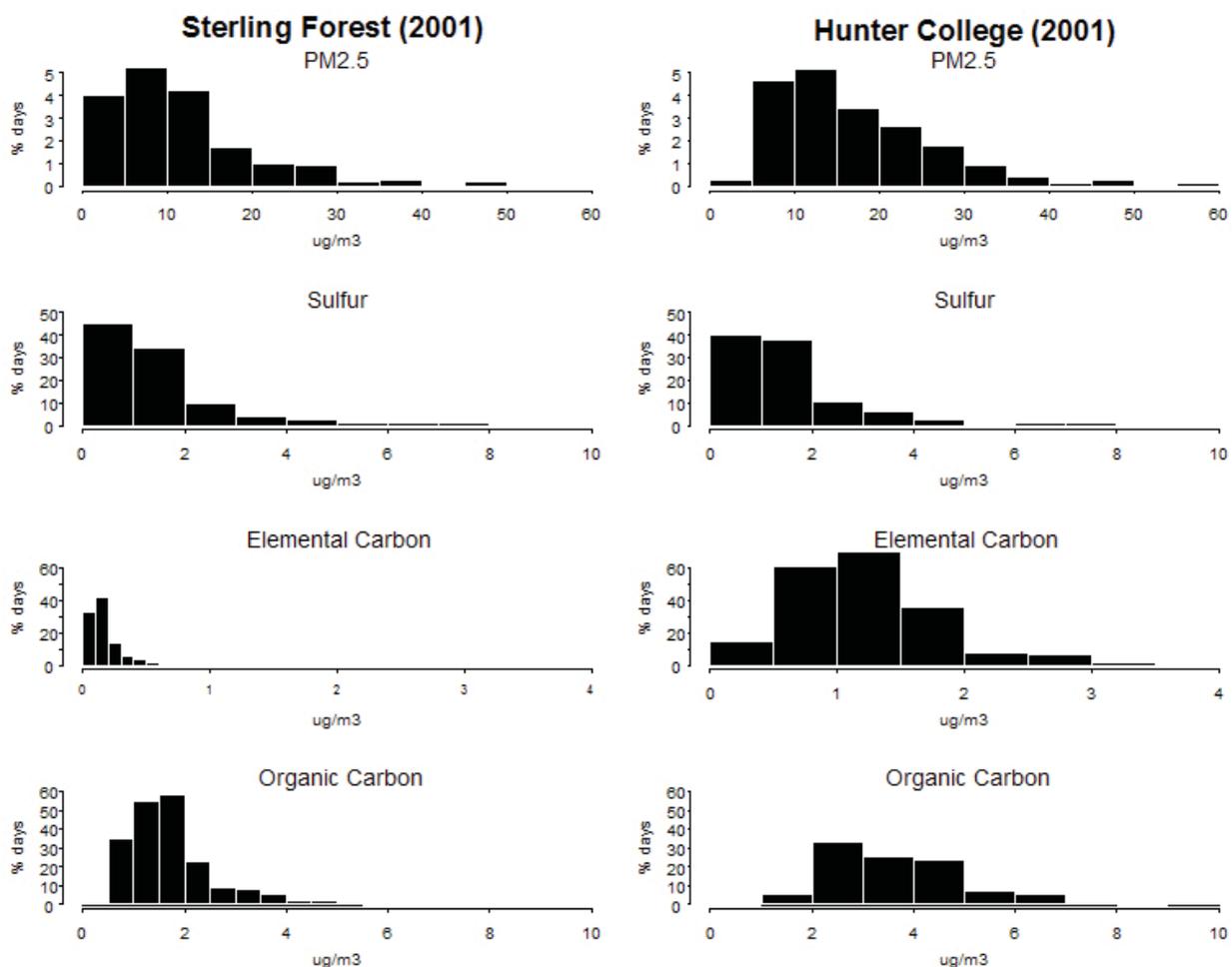
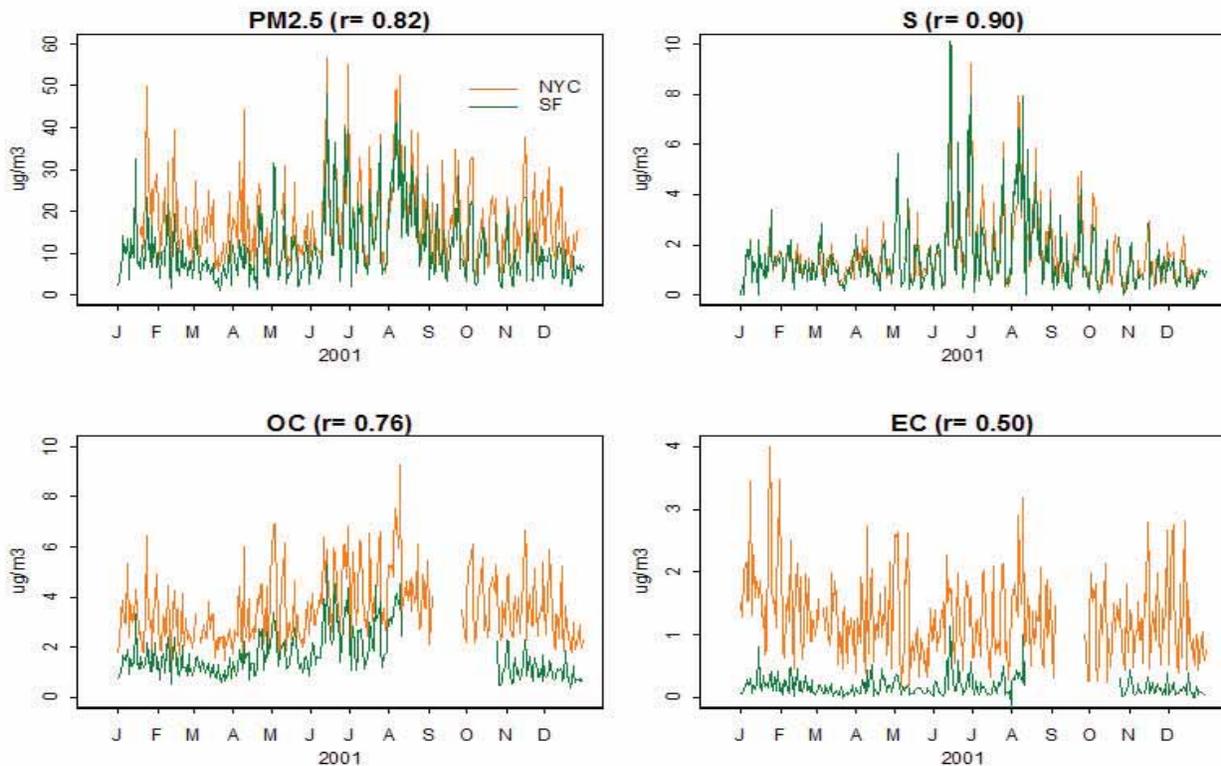


Table 1: PM_{2.5} mass concentration levels at NYU compared to other Manhattan DEC FRM monitors

	Site	PM _{2.5} mass (µg m ⁻³)
NYU	Hunter College (Sterling Forest)	17.30 (11.4)
NYDEC	JHS 45	15.20
NYDEC	PS 59	18.07
NYDEC	PS 59 (duplicate)	17.97
NYDEC	Canal Street	17.57
NYDEC	Canal Street (duplicate)	18.32

concentration average = 17.3 µg/m³) has comparable levels to other NYDEC sites in Manhattan (annual average of Manhattan sites = 17.1 µg/m³). However, Hunter College has much higher concentrations than Sterling Forest (annual average = 11.4 µg/m³). Further, as shown in the time-series plots in Figure 5, the PM_{2.5} mass concentrations at the two NYU sites are highly correlated

Figure 5: Time-series plots of PM_{2.5}, S, OC, and EC daily 2001 concentrations for NYC and Sterling Forest



($r = 0.82$). The linear regression of NYC $PM_{2.5}$ ACCU mass concentrations onto Sterling Forest ACCU mass concentrations has a slope of 0.94 and intercept of $6.6 \mu\text{g}/\text{m}^3$ over the whole year (Figure 6). But in the summer months, the slope rises to nearly 1:1 (0.99), and the intercept (i.e., NYC mass unexplained by pollution also affecting the Sterling Forest site) falls to $3.1 \mu\text{g}/\text{m}^3$ (Figure 6b). Given that Sterling Forest has few local pollution sources and is rarely impacted by pollution from NYC, this would suggest that, on an annual basis, up to two-thirds of New York City's fine PM pollution is a result of transported aerosols. Further, summer months (June-August) are associated with higher concentrations, as compared to winter months at both sites (summer: Hunter College = 21.4 vs. Sterling Forest = 18.3; winter: Hunter College = 17.3 vs. Sterling Forest = 8.8) (Figures 6a and 6b, and Table 2). Therefore, while in winter approximately 50% of NYC's $PM_{2.5}$ could be associated with background (Sterling Forest) $PM_{2.5}$, in the summer months some 85% of NYC's $PM_{2.5}$ appears to be associated with background transported aerosols.

Figure 6: A simple linear regression of NYC $PM_{2.5}$ mass onto Sterling Forest (SF) mass suggests that, on an annual basis, up to two-thirds of New York City's fine PM pollution is a result of transported aerosols.

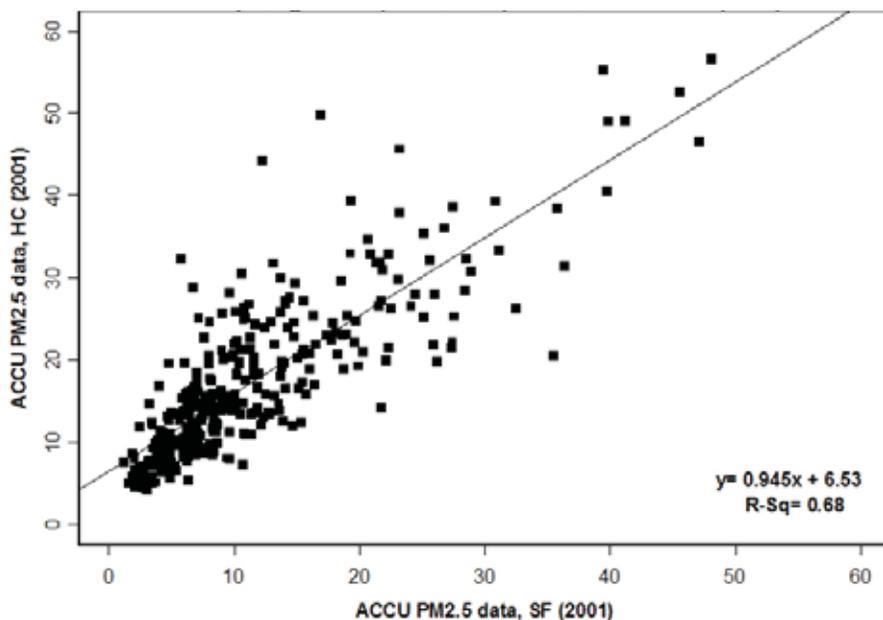


Figure 6a: Roughly half of NYC wintertime (Jan-Mar 2001) filter-based PM_{2.5} is well predicted by PM_{2.5} measured at the background Sterling Forest (SF) site.

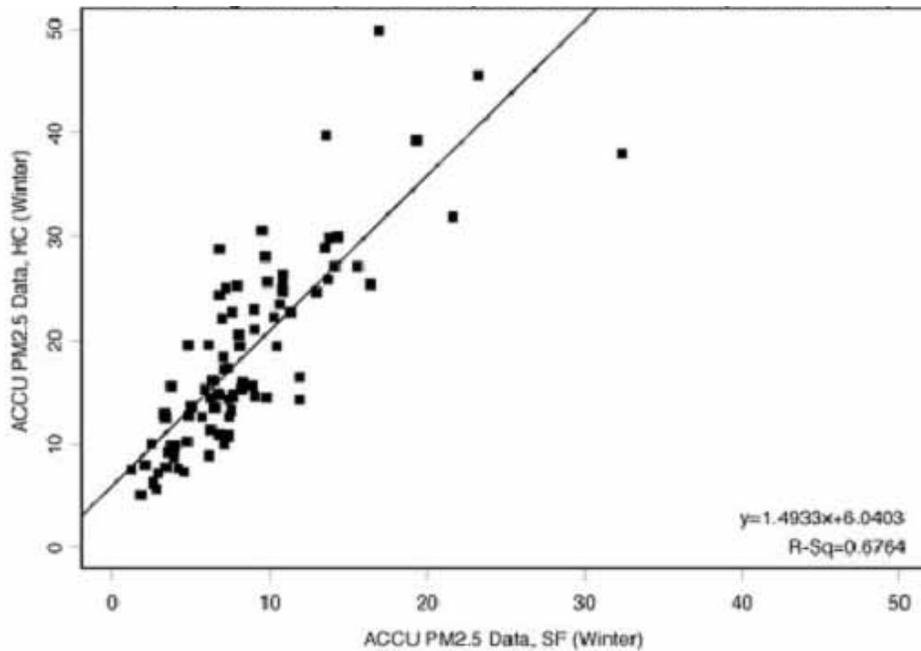


Figure 6b: Most NYC summer (Jun-Aug 2001) ACCU PM_{2.5} mass is well predicted by PM_{2.5} measured at the background Sterling Forest (SF) site.

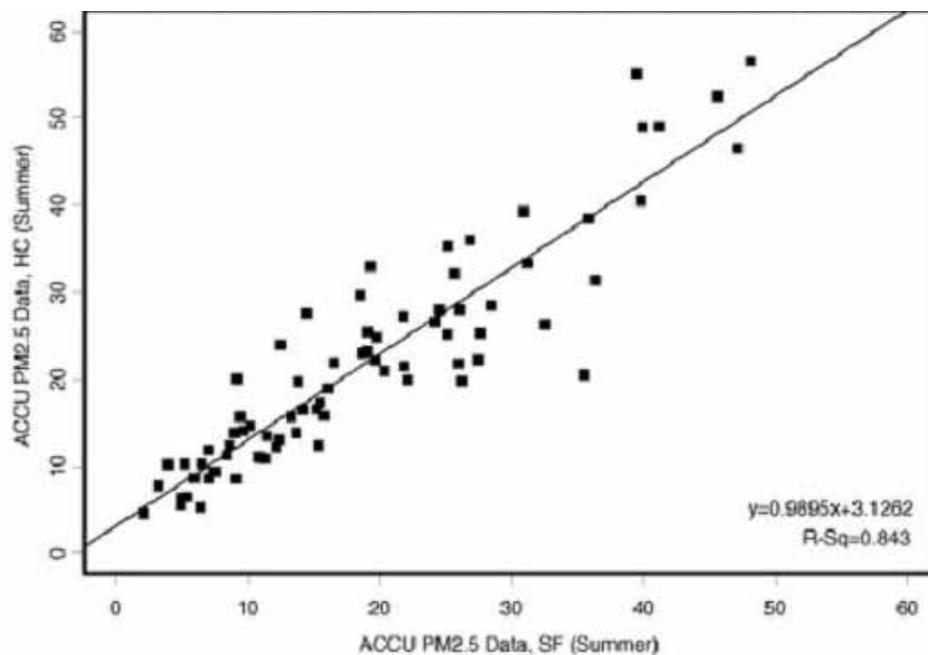


Table 2: Mean annual and seasonal concentrations (and their standard deviations) of EC, OC, S, and PM_{2.5} measured at the two NYU sampling sites during 2001 ($\mu\text{g}/\text{m}^3$)

	2001	Sterling Forest	New York City
Elemental Carbon	Annual	0.2 (0.2)	1.2 (0.6)
	Winter	0.2 (0.1)	1.3 (0.7)
	Summer	0.2 (0.2)	1.3 (0.6)
Organic Carbon	Annual	1.7 (1.0)	3.6 (1.3)
	Winter	1.3 (0.5)	2.7 (0.9)
	Summer	2.9 (1.3)	4.4 (1.3)
Sulfur	Annual	1.5 (1.4)	1.6 (1.3)
	Winter	1.1 (0.6)	1.2 (0.5)
	Summer	2.6 (2.2)	2.5 (1.9)
PM _{2.5}	Annual	11.4 (8.5)	17.3 (9.6)
	Winter	8.8 (5.4)	17.3 (9.0)
	Summer	18.3 (11.0)	21.4 (11.9)

Winter= January-March; Summer=June-August (instead of standard quarters).

Comparisons of the two NYU sites' elemental concentrations are also instructive as to the potential role of local versus transported pollution in NYC. Sulfur is a useful tracer for transported combustion aerosols in the northeastern United States (e.g., Lioy and Thurston, 1987). Sulfur, like PM_{2.5}, is found to be highly correlated between the two NYU sites ($r = 0.90$) (Table 3). Both sites are found to have similar sulfur levels (annual mean: Hunter College = 1.6 $\mu\text{g}/\text{m}^3$; Sterling Forest = 1.5 $\mu\text{g}/\text{m}^3$), with higher levels at both sites in the summer (Hunter College = 2.5, Sterling Forest = 2.6) compared to winter (Hunter College = 1.2, Sterling Forest = 1.1) (Table 2). On the other hand, elemental carbon (EC), which serves as a tracer of local combustion sources, is much less correlated between the two sites ($r = 0.50$). Since the EC concentrations are much higher in the city (mean = 1.2 $\mu\text{g}/\text{m}^3$) than in Sterling Forest (mean = 0.2 $\mu\text{g}/\text{m}^3$), most of the elemental carbon at the Hunter College site can be attributed to local sources (Table 2). There are no appreciable seasonal differences found for this variable (summer: Hunter College = 1.3, Sterling Forest = 0.2; winter: Hunter College = 1.3, Sterling Forest = 0.2).

Table 3: Number of observations used in the comparison between two sites (n), and the correlation of PM_{2.5}, sulfur, OC, and EC

	n	PM _{2.5}	n	S	OC	EC
SF vs. CH	88	0.75	50	0.95	0.69	0.33
HC vs. CH	87	0.80	52	0.96	0.74	0.52
SF vs. HC	312	0.82	312	0.92	0.70	0.49

SF=Sterling Forest (NY), NYC=New York City (NY), and CH=Chester (NJ).

The data in Figure 7 show that the difference between the Hunter College site $PM_{2.5}$ and the upwind Sterling Forest $PM_{2.5}$ is proportional to the elemental carbon concentration at the Hunter College site. This supports the hypothesis that the difference between the two sites is due to local combustion pollution, as well as the fact that elemental carbon is a reasonable marker of that local air pollution. A summary of the mean (and standard deviations) of the mass and elemental concentrations as well as the XRF method detection limits for each of the elements is provided in Table 4.

Figure 7: Comparing NYC EC to the difference in TEOM $PM_{2.5}$ values between NYC and Sterling Forest in winter (January-March) of 2001, indicating that elemental carbon is an excellent predictor of the local (non SF) wintertime $PM_{2.5}$

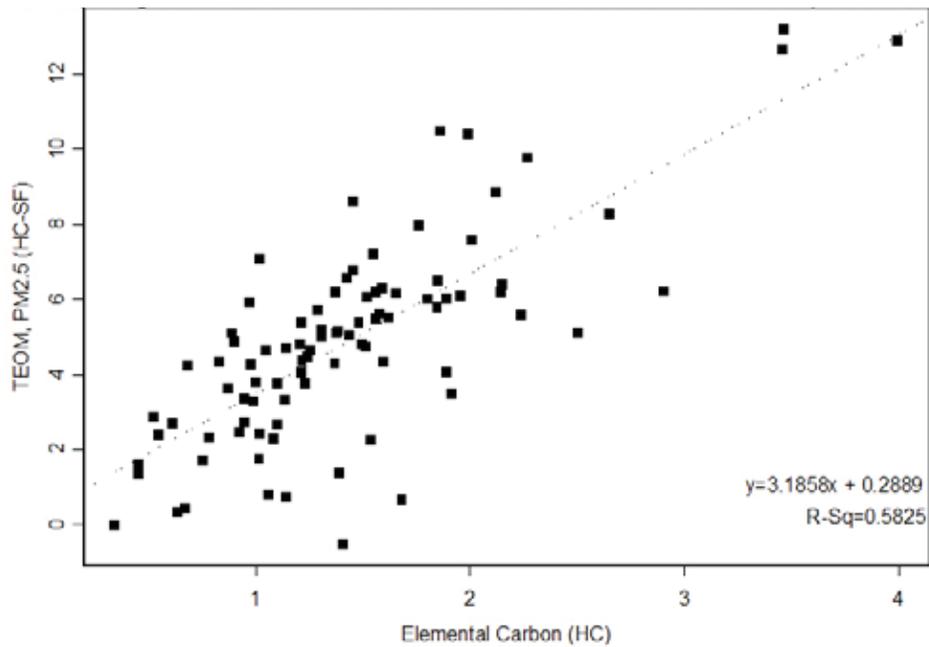


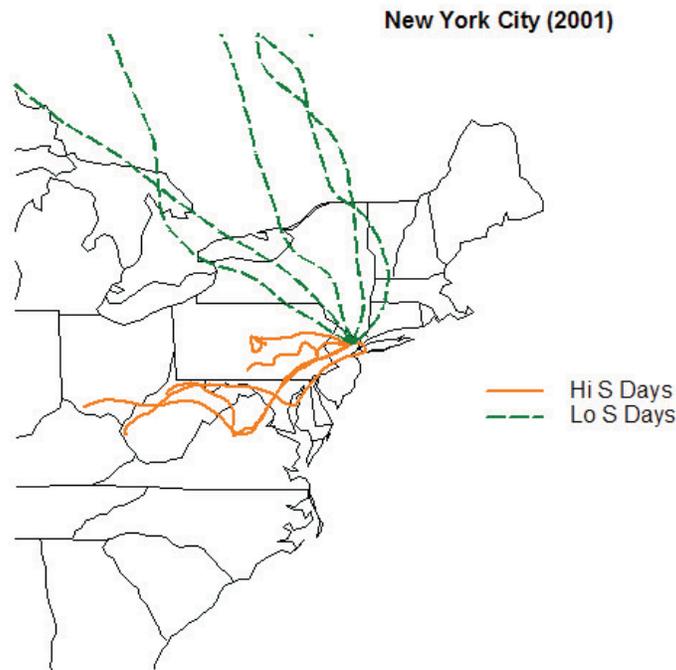
Table 4: Mean, standard deviation of PM_{2.5}, trace elements and carbon data (ng/m³) for the two NYU sites and the % above detection based on the XRF trace element detection limits

El	XRF DL (Teflon G, ng/m3)	Sterling Forest (SF)			New York City (NYC)		
		Mean (ng/m3)	Std. Deviation	% Above detection	Mean (ng/m3)	Std. Deviation	% Above detection
PM _{2.5}	-	11358	8301	-	17326	9642	-
Na	34	39	44	40	85	67	81
Mg	26	12	15	9	20	16	23
Al	64	42	57	17	44	50	18
Si	46	67	126	38	134	166	84
S	19	1512	1432	99	1583	1317	100
Cl	13	-2	11	4	37	273	30
K	12	36	27	92	50	49	99
Ca	8	20	19	80	60	36	100
Ti	6	4	18	7	4	4	16
V	4	3	3	27	10	6	88
Mn	5	1	1	1	7	11	31
Fe	5	39	40	95	194	131	100
Ni	4	4	14	23	24	14	98
Cu	4	1	2	10	6	13	47
Zn	3	9	7	88	44	97	100
Se	3	1	1	8	3	2	38
Br	4	3	9	24	8	37	74
Sr	3	1	1	1	3	3	27
Ba	11	0	3	0	9	5	31
Pb	12	2	5	2	9	36	17
OC	34	1700	856	-	3597	1260	-
EC	26	174	139	-	1226	580	-

Evaluating Sterling Forest as a NYC background site

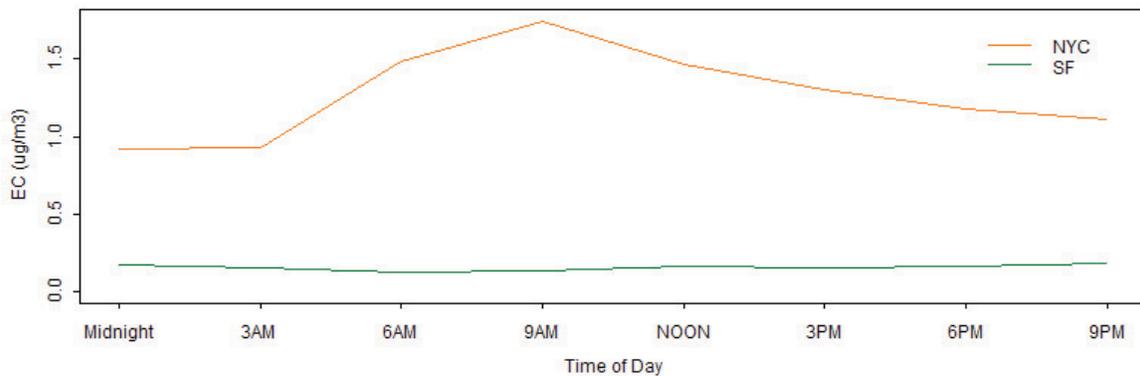
In addition to examining the New York City wind rose (Figure 3), which indicated that the winds rarely blow from the southeast toward the Sterling Forest site, several aerosol concentration-based approaches were examined to evaluate the appropriateness of using Sterling Forest as a background site for NYC (because it is affected by regionally transported pollution, but not by NYC's pollution). As shown in Figure 8, a review of the air mass back-trajectories on the highest and lowest sulfur pollution days in 2001 at the Hunter College site, as modeled using the NOAA Air Resources Laboratory (ARL) HYSPLIT model (Draxler, 1999), indicates there is consistency in the source regions of each group. The highest sulfates were consistently found to occur on days with air mass transport from the west and southwest, while the lowest sulfate days occurred during air mass transport from the north-northwest, both of which indicate that regional

Figure 8: Air mass back-trajectories for the five highest and five lowest fine particle sulfur concentration days in New York City, confirming the importance of regional transport to this component of PM_{2.5}.



transport is an important factor in defining the particulate sulfate concentrations at the Hunter College site. Conversely, an examination of the time of day concentration of elemental carbon at the Hunter College and Sterling Forest sites shown in Figure 9 indicates that the elemental carbon levels in the city are much higher than in Sterling Forest, and peak during the morning rush hour, indicating that the elemental carbon-related mass in NYC is largely of local origin. The 3-hour EC averages at Sterling Forest do not reveal a similar traffic pattern, which suggests that Sterling Forest does not have any significant local traffic sources and does not appear to be impacted by NYC local PM pollution.

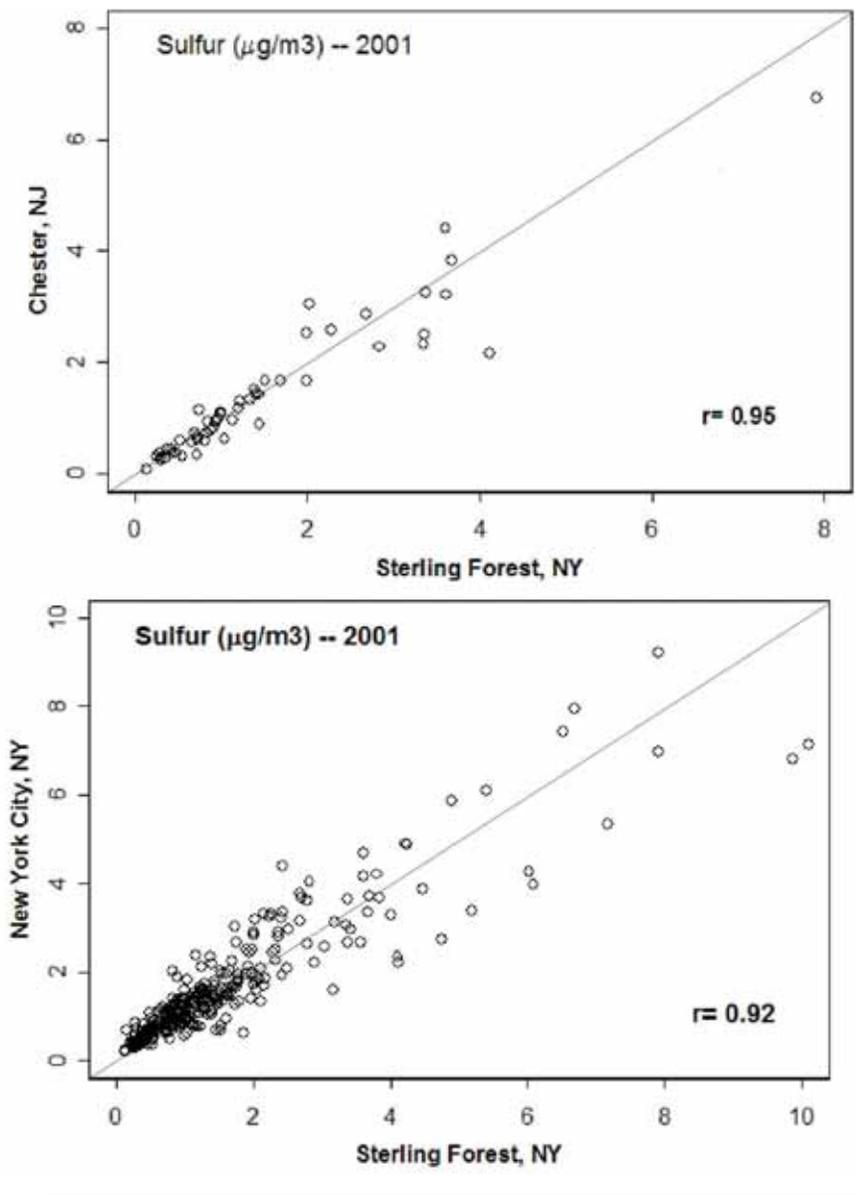
Figure 9: Comparison of 2001 average time-of-day elemental carbon (EC) concentrations at the Hunter College and Sterling Forest sites, indicating an NYC diurnal cycle consistent with a major role of traffic in NYC EC



To further test the suitability of Sterling Forest as a background site for the New York metropolitan area, the elemental carbon data collected at Sterling Forest were compared with both an STN “background” site located in Chester, NJ, and with the Hunter College site. It is apparent from the results in Table 3 that, while $PM_{2.5}$, sulfur, and OC are highly correlated across the three sites ($r > 0.9$ for S, $r > 0.7$ for OC), EC is far less correlated ($r < 0.6$). This indicates that OC and sulfate are useful tracers of regionally transported and secondary aerosols that affect all sites similarly (Figure 10), while EC is a tracer of more local combustion-related sources that vary more from site to site. The slight EC correlations found among the sites are likely primarily related to the three locations having similar local weather conditions (e.g., good versus poor dispersion characteristics) from day to day, rather than any shared pollution sources for EC. A table of the mean and standard deviations of the mass and elemental concentrations at Sterling Forest and Hunter College are provided in Table 4, and a summary of the $PM_{2.5}$, sulfur, organic carbon, and elemental carbon concentrations for the two sites by season are provided in Table 2.

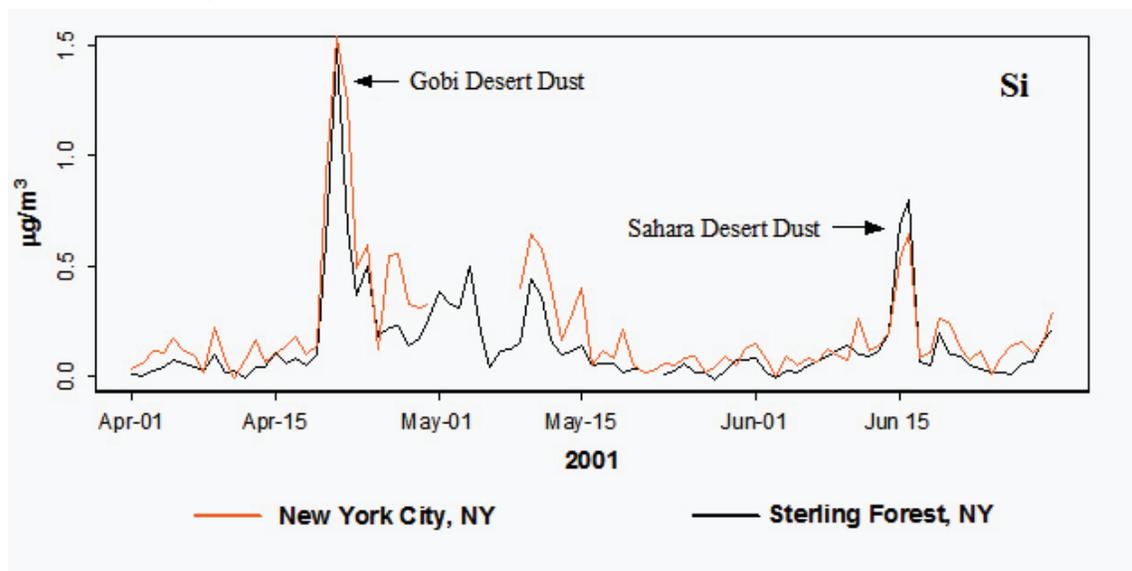
Our 2001 data also indicate that Sterling Forest is useful in verifying trans-continental and other transported aerosol episodes, like the Gobi Desert sandstorm dust transport episode of April 2001, which originated in Mongolia and traveled eastward around the globe and across the US from west to east at this time (US EPA, 2003b). An unusual peak was recorded at both our sites in April, as the result of air masses carrying dust particles that originated in a Gobi desert dust storm, and were transported across the Pacific Ocean to the West Coast of the United States and across the nation. This episode is well documented by the USEPA and the National Aeronautics and Space Administration (NASA), and the impact of this trans-continental long-range source is further confirmed by our data (USEPA, 2003b). In mid-June, a smaller regional dust peak was again observed at both our sites. Sequential satellite images taken during this period document a large plume originating in the Sahara Desert in Africa that migrated westward across the Atlantic Ocean in mid-June, 2001 (NASA, 2001). The episode recorded at our site on June 15-16 is due to an early precursor of the larger plume that mainly impacted the Caribbean and the Gulf of Mexico a few days later (June 19-21, 2001). Backward wind-trajectories for NYC for June 15-16 confirm winds blowing from over the Atlantic Ocean to NYC, suggesting that the winds might have intercepted the earlier parts of this large desert dust plume. Examination of satellite photos also confirm an early pulse of aerosol into the mid-Atlantic at that time. STN data for NYC do not reveal this episode, as samples were not collected on a daily

Figure 10: Sulfur measurements collected at the two background sites, Sterling Forest, NY, and Chester, NJ; and sulfur measurements collected at Sterling Forest versus Hunter College, NYC



basis at NYC STN sites, and it so happened that no STN samples were scheduled for collection on these particular days (June 15-16, 2001). Figure 11 shows a plot of silica at the two sites, and the two clearly identifiable peaks at both sites align with transported desert dust particles reported as being carried across from the Gobi desert and the Sahara Desert at those times, respectively. These events clearly demonstrate that transported aerosols other than sulfates are also similarly experienced at both the Sterling Forest and Hunter College sites under consideration by this research. Such clear and corresponding patterns between the two sites were

Figure 11: Comparison of the fine particle silica concentrations between the two NYU sites (Sterling Forest and Hunter College, NYC) during trans-continental desert dust episodes



not found for PM components that are routinely associated with local sources of PM pollution in NYC (e.g., elemental carbon or nickel).

PMF source apportionment analysis: separating transported and local sulfur in the model

The source apportionment analysis of NYC data that we conducted identifies six sources of $\text{PM}_{2.5}$, as shown in Table 5 (a,b). The incorporation of the Sterling Forest sulfur data into the PMF model helped to clearly define sources as being either transported or local. Two of the six sources, namely, “Transported Sulfates” and “Trans-Continental Desert Dust” are considered non-local sources. The categorization of the “Trans-continental desert dust” as non-local is due to the large plumes of soil particles contributing to this source, during two known episodes (as described earlier). The remaining four sources of “Residual Oil Burning,” “Traffic,” “Local Iron-Manganese Dust,” and “WTC Fires” are clearly defined as local NYC pollution sources. It should be noted that the PMF analysis of the single Hunter College site resulted in similar sources (results from this separate analysis are not shown here), but the differencing method employed here allows a more definitive interpretation of the sulfate factor as non-local.

Table 5 (a): Correlations between trace elements and carbon vs. the six sources identified for NYC (using two Sulfur variables in the PMF model); Annual and quarterly mass contributions ($\mu\text{g}/\text{m}^3$) for 2001 (95% CI of contribution estimates)

	TRANSP. SULFATES	TRAFFIC	RESID. OIL	TRANSP. DESERT DUST	Fe-Mn or "LOCAL" DUST	WTC
Na	0.18	-0.06	-0.07	0.37	0.41	0.18
Mg	0.13	0.09	-0.15	0.81	0.21	0.06
Al	0.11	0.12	-0.23	0.88	0.15	-0.18
Si	0.11	0.09	-0.12	0.88	0.19	0.29
Cl	-0.08	-0.08	0.07	-0.08	-0.04	0.91
K	0.15	0.08	-0.09	0.34	0.17	0.76
Ca	0.11	0.40	-0.14	0.78	0.35	0.09
Ti	0.13	0.11	-0.22	0.83	0.28	0.00
V	0.45	0.37	0.40	0.13	0.18	0.06
Mn	0.06	0.15	-0.16	0.02	0.99	0.05
Fe	0.12	0.36	-0.16	0.37	0.87	0.06
Ni	-0.09	0.01	0.90	-0.18	-0.13	-0.08
Cu	0.03	0.11	-0.04	-0.03	0.17	0.95
Zn	-0.02	0.08	0.03	-0.04	0.09	0.93
Se	0.58	0.40	0.09	-0.01	0.17	0.15
Br	-0.01	-0.08	-0.04	-0.03	-0.02	0.98
Sr	-0.02	-0.14	0.10	0.14	-0.08	0.72
Ba	0.23	0.43	-0.04	0.53	0.14	-0.02
Pb	-0.02	-0.03	0.01	-0.04	0.04	0.99
OC	0.65	0.82	-0.32	0.19	0.32	0.10
EC	0.39	0.80	0.20	0.10	0.17	0.10
S (TRANSP.)	0.92	0.44	-0.25	0.11	0.01	-0.02
S (LOCAL)	-0.20	0.12	0.09	-0.01	0.11	0.10
Eigen Value	2.21	2.42	1.43	4.31	2.53	5.86
2001 ANNUAL CONTRIBUTIONS [$\mu\text{g}/\text{m}^3$]	7.9 (7.4 - 8.4)	6.7 (5.9 - 7.5)	3.4 (3.0 - 3.9)	1.1 (0.8 - 1.4)	0.4 (0.1 - 0.6)	0.4 (0.4 - 0.5)
WINTER [JAN-MAR] CONTRIBUTIONS [$\mu\text{g}/\text{m}^3$]	6.4 (5.9 - 6.8)	4.8 (4.2 - 5.3)	7.7 (6.7 - 8.7)	0.5 (0.3 - 0.6)	0.2 (0.1 - 0.3)	0.3 (0.3 - 0.4)
SPRING [APR-JUN] CONTRIBUTIONS [$\mu\text{g}/\text{m}^3$]	8.7 (8.1 - 9.2)	6.4 (5.7 - 7.1)	2.4 (2.1 - 2.7)	2.1 (1.5 - 2.7)	0.2 (0.1 - 0.3)	0.2 (0.2 - 0.2)
SUMMER [JUL-SEP] CONTRIBUTIONS [$\mu\text{g}/\text{m}^3$]	10.5 (9.8 - 11.2)	8.3 (7.4 - 9.2)	0.9 (0.8 - 1.1)	0.8 (0.6 - 1.1)	0.4 (0.1 - 0.7)	0.8 (0.7 - 1.0)
FALL [OCT-DEC] CONTRIBUTIONS [$\mu\text{g}/\text{m}^3$]	5.3 (5.0 - 5.7)	6.8 (6.0 - 7.6)	3.7 (3.3 - 4.2)	0.9 (0.6 - 1.1)	0.8 (0.3 - 1.3)	0.4 (0.3 - 0.5)

Note: "Traffic" is all mobile vehicular (gasoline & diesel sources).

Table 5 (b): Source profiles of the six NYC sources from PMF-2 model (using two sulfur variables)

	TRANSP. SULFATES	TRAFFIC	RESID. OIL	TRANSP. DESERT DUST	Fe-Mn or "LOCAL " DUST	WTC
Na	0.2	0.0	0.4	1.3	4.6	0.4
Mg	0.0	0.0	0.0	0.9	0.5	0.1
Al	0.0	0.1	0.0	2.6	0.9	0.0
Si	0.0	0.2	0.2	6.1	4.0	1.5
Cl	0.0	0.0	0.1	0.0	0.0	0.2
K	0.1	0.1	0.2	1.3	0.8	1.7
Ca	0.0	0.3	0.1	2.3	1.7	0.2
Ti	0.0	0.0	0.0	0.2	0.2	0.0
V	0.0	0.0	0.1	0.1	0.1	0.0
Mn	0.0	0.0	0.0	0.0	1.6	0.0
Fe	0.1	0.9	0.4	3.9	16.8	0.3
Ni	0.0	0.1	0.4	0.0	0.0	0.0
Cu	0.0	0.0	0.0	0.0	0.2	0.8
Zn	0.0	0.2	0.2	0.1	0.1	4.3
Se	0.0	0.0	0.0	0.0	0.0	0.0
Br	0.0	0.0	0.0	0.1	0.0	0.6
Sr	0.0	0.0	0.0	0.1	0.0	0.1
Ba	0.0	0.1	0.0	0.2	0.1	0.0
Pb	0.0	0.0	0.0	0.0	0.0	1.8
OC	10.2	26.7	10.3	31.0	60.2	7.5
EC	0.9	11.4	6.3	5.3	8.3	4.0
S (TRANSP.)	14.7	1.8	0.0	7.5	0.2	1.4
S (LOCAL)	0.0	1.1	1.0	0.0	2.1	1.9

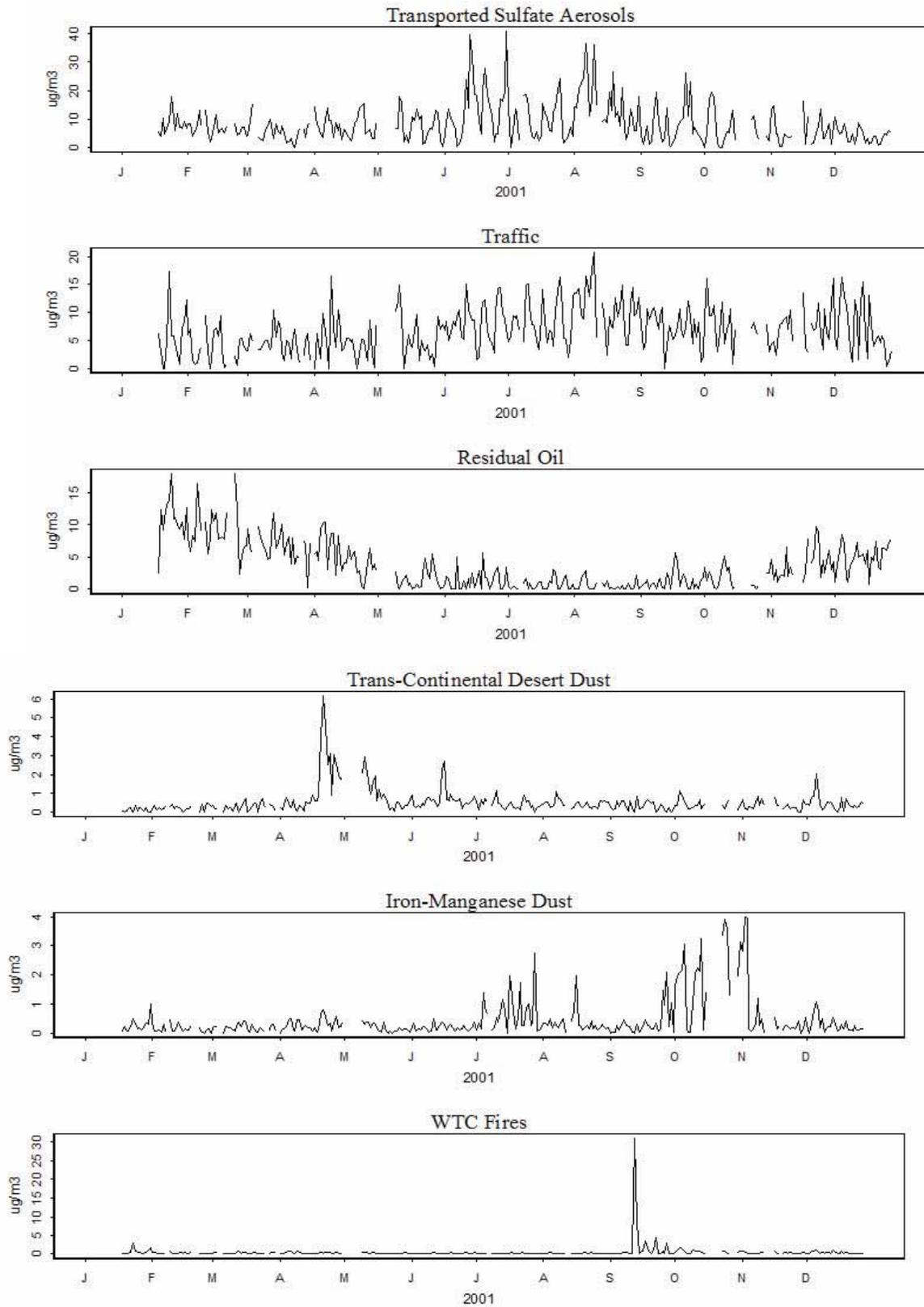
Table 5 (c): Average \pm std. error of PM_{2.5} concentrations and source contributions ($\mu\text{g}/\text{m}^3$) stratified by PM_{2.5} percentile-days

Percentiles	PM2.5	Transp. Sulfates	Traffic	Residual Oil	Transp. Desert Dust	Local Dust	WTC
0 - 5	6	1.3 \pm 0.3	3.2 \pm 0.5	2.3 \pm 0.5	0.6 \pm 0.1	0.1 \pm 0.0	0.1 \pm 0.0
5 - 10	7	1.7 \pm 0.5	5.2 \pm 0.8	2.7 \pm 0.7	0.7 \pm 0.1	0.1 \pm 0.1	0.0 \pm 0.0
10 - 20	9	3.5 \pm 0.4	4.3 \pm 0.4	2.7 \pm 0.4	1.0 \pm 0.2	0.2 \pm 0.1	0.1 \pm 0.0
20 - 30	11	3.8 \pm 0.4	4.9 \pm 0.4	3.5 \pm 0.5	0.8 \pm 0.1	0.4 \pm 0.1	0.2 \pm 0.0
30 - 40	13	5.8 \pm 0.7	4.8 \pm 0.6	3.2 \pm 0.6	1.1 \pm 0.2	0.5 \pm 0.2	0.2 \pm 0.0
40 - 50	15	6.1 \pm 0.5	5.7 \pm 0.4	3.8 \pm 0.5	0.8 \pm 0.1	0.4 \pm 0.1	0.4 \pm 0.1
50 - 60	17	6.6 \pm 0.6	5.7 \pm 0.7	4.9 \pm 0.5	1.0 \pm 0.2	0.4 \pm 0.1	0.3 \pm 0.0
60 - 70	21	9.0 \pm 0.6	7.6 \pm 0.6	3.3 \pm 0.7	1.5 \pm 0.3	0.6 \pm 0.2	0.4 \pm 0.1
70 - 80	25	10.3 \pm 0.8	8.2 \pm 0.6	3.2 \pm 0.7	1.6 \pm 0.4	0.4 \pm 0.1	0.6 \pm 0.3
80 - 90	30	12.0 \pm 1.1	9.7 \pm 0.7	4.4 \pm 0.7	1.3 \pm 0.4	0.3 \pm 0.0	0.5 \pm 0.1
90 - 95	35	16.3 \pm 1.9	10.9 \pm 1.1	2.2 \pm 0.8	1.4 \pm 0.3	0.7 \pm 0.2	3.2 \pm 2.2
95 - 100	57	23.9 \pm 2.7	13.0 \pm 1.2	3.2 \pm 1.2	0.9 \pm 0.2	0.4 \pm 0.1	0.5 \pm 0.2

Table 5 (d): Average source contribution estimates \pm std. error ($\mu\text{g}/\text{m}^3$) on weekdays vs. the weekend

mean \pm s.e. (ng/m3)	Transp. Sulfates	Traffic	Residual Oil	Transp. Desert Dust	"Fe-Mn" Local Dust	WTC Fires
Weekend	7.9 \pm 0.7	3.8 \pm 0.3	3.2 \pm 0.3	1.0 \pm 0.2	0.3 \pm 0.1	0.2 \pm 0.1
Weekdays	7.9 \pm 0.5	7.8 \pm 0.3	3.5 \pm 0.2	1.1 \pm 0.1	0.4 \pm 0.0	0.5 \pm 0.2

Figure 12: Time-series plots of the PM_{2.5} contributions ($\mu\text{g}/\text{m}^3$) from six source categories identified for NYC



Time-series plots of the mass contributions from each source are provided in Figure 12. In this analysis, total NYC PM_{2.5} mass concentrations were regressed onto the six factors, in order to translate factor scores into these mass contributions from each source:

$$\text{NYC PM}_{2.5} = \beta_0 + \beta_1 * G_{\text{TRANSPORTED}} + \beta_2 * G_{\text{TRAFFIC}} + \beta_3 * G_{\text{RESIDUAL OIL}} + \beta_4 * G_{\text{SOIL}} + \beta_5 * G_{\text{FE-MN}} + \beta_6 * G_{\text{WTC}},$$

where Gs are the factor scores of the component, and the β s are the regression coefficients for the regression of mass onto the factor scores.

Using the Sterling Forest sulfur concentrations in the NYC source apportionment analysis clearly indicates that the “Transported Sulfates” are truly being transported into the NYC area, a phenomenon documented by several previous studies (see description of these studies in the introduction). Wind trajectories on the highest days from this source were also useful in indicating this source as being primarily from the midwestern United States, around the Ohio Valley region (similar to Figure 8), where numerous coal-fired power plants are still operated, many still without sulfur-oxide control technology. This “source” is highly correlated with the “Transported Sulfur” component, organic carbon, and selenium (a reliable tracer of coal burning in the eastern United States). Approximately 15% of particle mass from this source consists of sulfur (see source profile in Table 5(b)), which would mostly be in the form of sulfuric acid and ammonium bisulfate. On an annual basis, and especially during the summer months, this source is the largest contributor to the PM_{2.5} mass (approximately 45% and 56%, respectively). Due to summertime photochemistry, the time-series plot for this source displays a very distinct seasonal pattern, with the highest concentrations occurring during the period from June through August (summer = 10.5 $\mu\text{g}/\text{m}^3$ vs. annual = 7.5 $\mu\text{g}/\text{m}^3$). On several days during this period, concentrations from this source alone exceed 35 $\mu\text{g}/\text{m}^3$ (i.e., the newly revised PM_{2.5} 24-hour standard). In general, high PM_{2.5} days are also correlated with high transport aerosol days (see Table 5(c)).

The “Trans-Continental Desert Dust” component profile has high percentages of elements associated with the earth’s crust (e.g., Al and Si). As shown in Figure 11, there are two distinct peaks found in the time-series plots that relate to the Gobi Desert dust and Sahara Desert dust episodes, respectively. However, there also appear to be smaller contributions year-round,

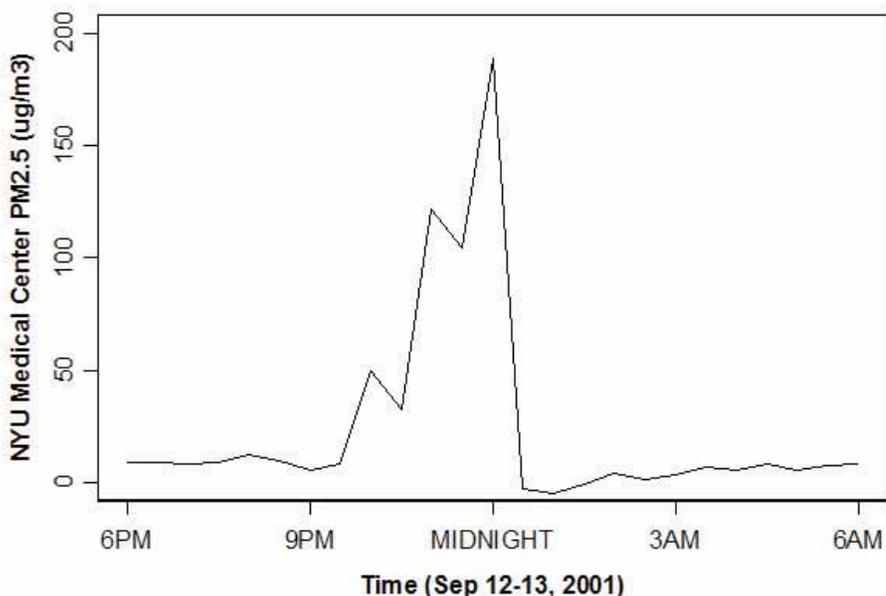
which cannot necessarily be attributed to transport. Since elements associated with these storms are similar to those measured in NYC on a daily basis, both local and non-local contributions have likely aligned on the same “source” in this case. However, since PM_{2.5} levels associated with the dust storms are much higher than usual (more than 5 times higher than normal year-round levels: Gobi = 6.6 µg/m³, and Sahara = 5.1 µg/m³), this factor has been categorized as transported aerosols.

The “Traffic” and “Residual Oil” sources are found to be major local contributors to NYC PM_{2.5}. Even on relatively “clean” days (PM_{2.5} between 7 - 11 µg/m³) contribution from the traffic and residual oil sources are approximately 4-5 µg/m³ and more than 2.7 µg/m³, respectively (Table 5(c)). The “Traffic” component (i.e., mobile sources) is a mixture of elemental carbon and organics from vehicular exhaust and traces of re-suspended road dust including elements like Fe and Ca (Table 5(a, b)). This component also has a strong day-of-week pattern, with much lower concentrations on the weekend (mean = 3.8 µg/m³) compared to weekdays (mean = 7.8 µg/m³) (Table 5(d)), since traffic in NYC is much heavier on weekdays. The lowest 5th percentile of traffic contributions (< 0.9 µg/m³) coincided with Sundays mostly, and a few with Saturdays and holidays (Christmas and December 26). Elemental Carbon is a useful indicator of traffic in NYC, as is shown in Figure 9, where elevated levels in the every 3-hour EC measurements are found to coincide with traffic rush-hour peaks. Residual oil is used by power plants and NYC apartment building boilers, and could potentially include emissions from NY/NJ ports, where large ships burn “bunker” fuel. This component’s elemental profile includes nickel and vanadium, two tracers commonly associated with this type of fuel. Much of the “Residual Oil” in this analysis (as shown in the time-series plot) is found to occur during the colder months (winter = 7.7 µg/m³ vs. summer = 0.9 µg/m³), when there is a greater demand for indoor space-heating. In fact, between January and March this source is the largest contributor to total PM_{2.5}. As expected, this residual oil combustion source is found to be highly correlated with EPA’s daily NYC ambient SO₂ data for this period (r = 0.58).

The two other local sources identified as “WTC” and “Local Fe-Mn Dust,” unlike traffic, are not year-round events and seldom occur, and therefore are to be treated as infrequent episodes rather than regular sources. The WTC plume is a result of the destruction and fires following the attack on the WTC towers. This plume, laden with chlorine, zinc, lead, copper, and potassium particles, hit the Hunter College site between 9pm and midnight on September 12th,

according to the TEOM records (see Figure 13). The iron-manganese source most likely is a result of re-suspended dust that is local in origin, and is therefore labeled as “Local Dust.” However, this ambiguous source category only constitutes less than $0.5 \mu\text{g}/\text{m}^3$ on average. An increase in levels is found in the dryer months (from July to early August) and again for a period between late September and mid-November. These elevated levels toward the latter part of the year might reflect the re-suspension of WTC-related dust during the Ground Zero cleanup from October-November 2001.

Figure 13: The WTC plume hit the Hunter College site on September 12, 2001, between 9pm and 12am



DISCUSSION

Since the 1970s, multiple studies have been published relating to the air quality of the New York metropolitan region and, in particular, to the impact of sulfate aerosols transported into this region. Wind-trajectory analyses were conducted in some of these studies, allowing them to identify coal-fired plants in the Ohio River Valley as a major cause of the sulfates measured in NYC. Indeed, one early study estimated that some 73% of sulfate measured in the New York City metropolitan area is attributed to pollution transport (Lippmann, 1979). In addition to identifying high sulfur concentrations originating from emissions occurring in the midwestern United States, another study was also able to show meteorological conditions

conducive to the transport of this pollution into New York State (Galvin, 1978): stagnant air masses over the Ohio River Valley were found to follow the movement of the high-pressure system eastward into New York State. On such days, when New York experienced winds from around the industrialized region located to its west and southwest, sulfate concentrations in excess of $20 \mu\text{g}/\text{m}^3$ were recorded (versus approximately $5 \mu\text{g}/\text{m}^3$ on days with wind trajectories from other directions).

In the three decades since the above-mentioned studies, there have been some major reductions in the emissions of SO_2 and primary particulate matter in the eastern United States, which have therefore resulted in a decrease in the atmospheric burden of particulate matter trace elements and an overall improvement in air quality (Husain et al., 2004). Despite these reductions, long-range transported aerosols can still play a significant role in NYC's air quality, and, on a few days during the summer months, concentrations greater than $20 \mu\text{g}/\text{m}^3$ are still occasionally observed. More recent studies estimate about 40-60% of the sulfates, and approximately 30% of the total $\text{PM}_{2.5}$, are due to transport from upwind sources (e.g., Dutkiewicz et al., 2004; Bari et al., 2003). These estimates are lower than the ones found in this study. However, these two other studies used concentration data stratified by wind directions in order to estimate contributions from transported air (rather than the quantitative source apportionment techniques used in this study). This difference in assumptions may account for these differences in results if, for example, the wind from "reference" directions also include some transported pollution, and pollution experienced during those winds were not entirely local in origin.

Similar PM data collected at other NYC metropolitan area sites since 2000 (through EPA's STN) have been used in several recent source apportionment studies. These studies provide a more comprehensive evaluation of the multiple and varied sources that impact air quality in general and, along with supporting meteorological data, they allow an assessment of the impacts from these different sources (although the STN data are usually collected less frequently than the daily data collected in this NYSERDA study). Two such source apportionment studies have been recently conducted using available STN data for multiple sites in the NYC metropolitan area (Ito et al., 2004; Qin et al., 2006). Unlike, the NYU sampling schedule, the USEPA network only sampled every three days, and data are often not available on key days when high concentrations were recorded at the daily NYU sites, such as the result of an episode or unusual event (e.g., plumes of dust that originated from the Sahara Desert or as a

result of the destruction of the WTC by the 9-11 terrorist attacks). In both studies, separate and individual source apportionment analyses were conducted for each of the sites used in evaluating NYC's air quality. Ito et al. used three USEPA sites located in NYC boroughs outside Manhattan (i.e., the NY Botanical Gardens, I.S.52, and Queens College) and found sources (secondary aerosols, soil, traffic, and residual oil) similar to this study. Qin uses two additional sites located in Elizabeth, NJ, and Chester, NJ (a rural background site). Among the sources identified in that work, those found common to all five USEPA sites were: secondary sulfate, secondary nitrates, soil, and aged sea salt. Oil burning and a traffic source were also identified for certain of the sites. By comparing the actual mass and sulfate concentrations measured across the various NYC sites and a background NJ site, it was estimated that about 69-82% of $PM_{2.5}$ and more than 93% of the sulfates were attributed to transport. These findings are also consistent with our results. However, we have taken a slightly different approach in our analysis of speciation and carbon data—one that directly derived a separate estimate of the transported sulfate by subtracting concentration data from the background Sterling Forest site prior to the PMF source apportionment. This approach resulted in less ambiguity between contributions from local and distant sources and should provide a more specific assessment of transported versus local contributions to air quality in NYC. This may well aid regulatory agencies in adopting different strategies that can better target reductions in $PM_{2.5}$ sources at the city and regional levels.

Historically, past source apportionment studies have not considered incorporating an additional reference site in a site's source apportionment analysis in order to help separate local versus non-local sources affecting the air quality at an urban sampling site. However, this approach of incorporating background data was also extended by us to all 22 elements (instead of only sulfur data) used in the source apportionment analyses (Lall and Thurston, 2006). In order to provide additional insight into our use of this differing approach, we have also conducted a "conventional" PMF source apportionment analysis of just the NYC elemental dataset, without incorporating the Sterling Forest data. Although the results are not included in this report, this more conventional approach provided conclusions qualitatively and quantitatively similar to those presented here (i.e., resolving similar sources and comparable source contributions). These conventional PMF apportionment results were also similar to those reported in the work of Ito et al. (2004) and Qin et al. (2006) for other NYC sites. However, the approach in this study using background Sterling Forest sulfur concentrations as a tracer for the

transported component provides greater certainty that the sulfate component identified in the analyses of NYC data was, indeed, due to non-local aerosols transported into NYC. The validity of the results presented here is further supported by the fact that the PMF was able to correctly discern two very clear and well-documented episodes as distinct PM_{2.5} sources: the impact of the WTC fires plume in mid-Manhattan on September 12, 2001, and the Gobi Desert and Sahara Desert sandstorm inter-continental dust transport episodes in April and June, 2001, respectively.

Wind-rose plots using NOAA meteorological 2001 data show that, on a majority of study days, winds in NYC predominantly originated from the west, and very rarely did winds originate from the south-southeast direction (i.e., traveling in the direction from NYC toward Sterling Forest: see Figure 3). Back-trajectories of winds on days with the highest sulfur concentrations were found to indicate that the air mass originated in the midwestern United States. In contrast, on the cleanest days (in terms of sulfur concentrations), winds were from the north-northwest (see Figure 8). Sulfur concentrations measured at Sterling Forest, located to the northwest of NYC, were also compared with data from the USEPA background site located at Chester, NJ, also to the west of NYC. Similar concentrations were observed at both sites, and the sulfate data from these two sites were also highly correlated ($r = 0.95$). The same was found to be true when comparing sulfur concentrations between the two NYU sites. Such clear site-to-site patterns were not observed for elemental carbon (i.e., a component attributed to local combustion sources) concentrations at the two NYU sites. In general, aside from sulfur, higher concentrations of PM_{2.5}, trace elements, and carbon were usually found in Manhattan versus Sterling Forest. These facts validate the use of Sterling Forest as a reference site for our study.

The 2001 annual estimate of PM_{2.5} mass concentrations ($17.3 \mu\text{g}/\text{m}^3$) for NYC was above the federal annual standard ($15.0 \mu\text{g}/\text{m}^3$). This research, like other previous studies, also found transported PM_{2.5} and sulfate aerosols to have major impacts on NYC's air quality. Comparing PM_{2.5} mass and its components between the NYU sites on an annual basis, about 45% of the total PM_{2.5} measured in NYC is attributable to sulfate aerosols being transported into the city; during the summer months, the fraction due to transport can be as much as two-thirds (Table 6). For sulfates, the transported percentage is over 90% of all the sulfates affecting the Hunter College site in downtown Manhattan. By contrast, elemental carbon is mainly attributed to local pollution sources (95%). However, organic carbon is attributed to both local (~70%) and non-local (~30%) pollution. Nitrate data were not analyzed for the Hunter College samples (due to the generally

poor measurement of this pollutant), but the nitrate-related PM_{2.5} mass would be distributed among the local residual oil and transported aerosol components by the source apportionment model. Because the major contributions to total PM_{2.5}, sulfates and OC are attributed to transport in this model, it is indicated that, if NYC is to come into compliance with federal PM_{2.5} standards, it will need to specifically target regional PM_{2.5} reduction strategies. Along with alleviating PM_{2.5} concentrations in the northeastern United States and Canada, these strategies would also have the secondary benefits of improved visibility and reduced acid deposition in the sensitive ecological areas in this region.

Table 6: Estimated fractions of total PM_{2.5} mass and elemental/carbon concentrations contributed by transported aerosols in NYC

	ANNUAL (2001)	WINTER (Jan-Mar '01)	SUMMER (Jun-Aug '01)
PM _{2.5}	0.44	0.33	0.56
S	0.80	0.79	0.85
OC	0.31	0.26	0.67
EC	0.12	0.08	0.15

Overall, this study finds that the highest PM_{2.5} days occur when contributions from transported sulfate aerosols are highest (see Table 5(c)). In fact, on a few days in June and August, concentrations from this “source” alone were over 35 µg/m³ (i.e., the current revised 24-hour PM_{2.5} standard). Aside from the transported sulfate aerosols, two other, more local sources were found to contribute significantly: traffic and residual oil. “Cleaner” days (i.e., the 10th percentile of PM_{2.5} mass concentrations or less than 7 µg/m³), had greater contributions from the traffic and residual sources, compared to the transported sulfates. Therefore, the contributions of these local sources cannot be overlooked. In this work, no major contributions from WTC-related fires or rescue/clean-up activities at Ground Zero were distinctly identifiable, other than the direct impact of the plume from the WTC fires measured on September 12th at the Hunter College site.

Current PM-health studies indicate the significant contributions from combustion sources, and several studies find cardiovascular morbidity and mortality outcomes to be associated with traffic-related exposures (Laden, 2000; Hoek, 2002). NYC has previously

adopted policies to modernize the public transportation fleet to less-polluting vehicles (e.g., replacing diesel with cleaner hybrid buses). Current proposals for congestion pricing are being recommended to limit vehicles inside Manhattan's Central Business District (CBD). Such proposals would result in lower exposures to traffic; however, it is less certain how much these policies would also benefit the other NYC boroughs. Further investigation of the potential contribution to the residual oil category by emissions from ships at the NY/NJ ports is also necessary for a clearer understanding of the local $PM_{2.5}$ exposures in NYC.

This study used a new approach to source apportionment, subtracting elemental mass from a reference site prior to its statistical source apportionment analysis as a means of more clearly separating local versus upwind sources of $PM_{2.5}$. Air pollution at urban sites is a complex mixture of aerosols produced locally as well as those transported from great distances. This complicates the discernment of sources, especially when much of the mass is transported regionally. Using additional data from a nearby reference site (largely unaffected by local pollution sources) as an indicator of transported pollution is seen in this work to be a helpful approach to more clearly identify local versus upwind contributions to $PM_{2.5}$.

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