

Development of Advanced Factor Analysis Methods for Carbonaceous PM Source Identification and Apportionment



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INTRODUCTION

Because of controls on precursor gases that lead to sulfate and nitrate formation, carbonaceous particles are becoming a larger fraction of the fine particle aerosol. Accurate source identification and apportionment will be important for developing effective control strategies for areas found to be out of attainment of the PM_{2.5} standard. In addition, there is increasing interest in epidemiological studies to relate adverse health effects to apportioned source contributions. Thus, the objective of this project is to combine the best features of the two advanced factor analysis models, UNMIX and Positive Matrix Factorization (PMF), and to test the effectiveness of this improved factor analysis methodology by analysis of the data developed in the various supersites with an emphasis on data from the New York City supersite and other data from New York State. The NYC results are complete and presented below. Data from other sites are currently being analyzed.

EXPERIMENTAL SECTION

New York City Area

The locations of five STN sites in the metropolitan area of New York City are shown in Figure 1. The New York Botanical Garden site (NYBG) (40° 51' 58"N, 73° 52' 50"W) and the Intermediate School 52 site (IS52) (40° 48' 57"N, 73° 54' 07"W) are located in Bronx County, New York. The Queens College site (QCII) (40° 44' 11"N, 73° 49' 23"W) is located in Queens County, New York. The Elizabeth site (ELIZ) (40° 38' 28"N, 74° 12' 28"W) is located in Union County, New Jersey. All four sites were placed in urban commercial areas. The Chester site (CHES) (40° 47' 14"N, 74° 40' 31"W) is located in a suburban area in Morris County, New Jersey, about 100 Km west of New York City. The linear distance between IS52 and the other four sites is about 6, 11, 45 and 100 Km for NYBG, QCII, ELIZ and CHES, respectively. Details of the sampling and analysis processes are presented by Qin *et al.* (2005). The data consist of concentrations for PM_{2.5}, forty-eight elements by XRF, five ions by ion chromatography and organic (OC) and elemental (EC) carbon via the NIOSH protocol thermal optical transmission (TOT) method.

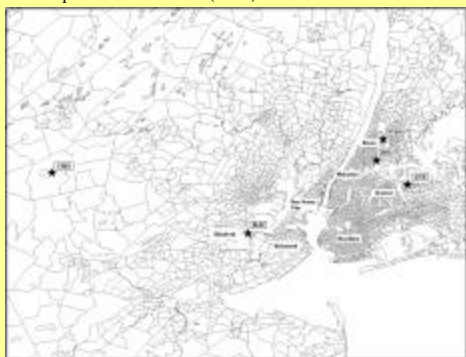


Figure 1. Map of New York City area showing the locations of the STN sites from which data have been analyzed.

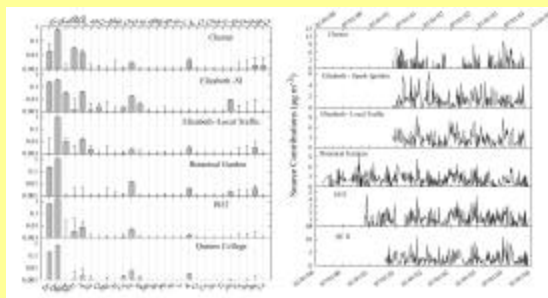


Figure 2. Source profiles and contributions for the spark-ignition vehicle emissions in the NYC area.

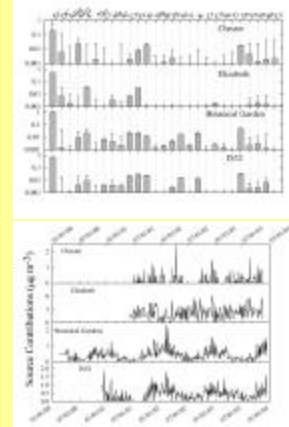


Figure 3. Source profiles and contributions for diesel vehicle emissions in the NYC area.

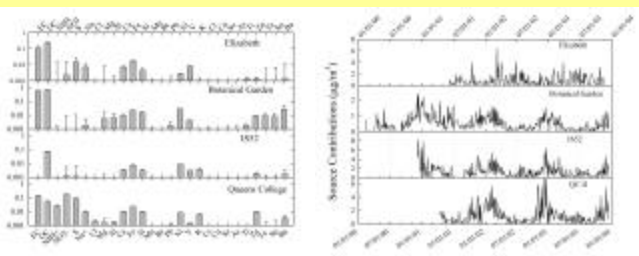


Figure 4. Source profiles and contributions for residual oil burning in the NYC area.

Table 2. Primary emission sources of PM_{2.5} in Bronx, Queens, Union and Morris counties (Tons mile⁻² year⁻¹) as given in the EPA Emissions Inventory

| Point | Bronx | Queens | Union | Morris | | | | |
|----------------|-------|--------|-------|--------|-------|-------|------|-------|
| | 0.89 | 1.3% | 9.03 | 13.9% | 3.18 | 9.7% | 0.14 | 1.7% |
| Area | | | | | | | | |
| Coal burning | 1.32 | 1.9% | 1.09 | 1.7% | 0.05 | 0.2% | 0.01 | 0.1% |
| Gas burning | 0.02 | 0.0% | 0.02 | 0.0% | 0.41 | 1.3% | 0.2 | 2.4% |
| Oil burning | 0.85 | 1.3% | 0.96 | 1.5% | 0.34 | 1.0% | 0.11 | 1.3% |
| Wood burning | 4.51 | 6.6% | 10.87 | 16.7% | 5.66 | 17.3% | 1.64 | 19.7% |
| Waste disposal | 3.17 | 4.7% | 2.49 | 3.8% | 3.48 | 10.6% | 1.1 | 13.2% |
| Fugitive dust | 38.65 | 56.9% | 24.86 | 38.2% | 12.85 | 39.2% | 3.42 | 41.2% |
| Other | 5.33 | 7.8% | 3.59 | 5.5% | 4.63 | 14.1% | 0.61 | 7.3% |
| On-Road | | | | | | | | |
| Gasoline | 3.06 | 4.5% | 1.91 | 2.9% | 0.78 | 2.4% | 0.16 | 1.9% |
| Diesel | 4.99 | 7.3% | 3.11 | 4.8% | 1.27 | 3.9% | 0.35 | 4.2% |
| Off-Road | | | | | | | | |
| Gasoline | 0.52 | 0.8% | 0.65 | 1.0% | 0.03 | 0.1% | 0.21 | 2.5% |
| Diesel | 4.65 | 6.8% | 6.51 | 10.0% | 0.07 | 0.2% | 0.36 | 4.3% |
| | 67.96 | 100% | 65.09 | 100% | 32.75 | 100% | 8.31 | 100% |

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Table 1. Average contributions of identified sources to PM_{2.5} concentrations

| | NYBG | ISS2 | QCII | ELIZ | CHES |
|-------------------|-----------|-----------|-----------|-----------|-----------|
| Secondary sulfate | 5.77±5.26 | 7.20±5.88 | 4.87±4.47 | 6.67±6.89 | 6.43±6.71 |
| Secondary nitrate | 2.10±2.45 | 2.57±2.44 | 1.81±2.55 | 1.98±2.38 | 1.05±1.41 |
| Soil dust | 1.48±1.06 | 1.07±0.76 | 0.75±0.64 | 0.99±1.41 | 0.99±0.78 |
| Aged sea salt | 0.68±0.69 | 0.49±0.61 | 0.44±0.61 | 1.14±1.37 | 0.87±1.74 |
| Oil combustion | 0.52±0.43 | 1.36±1.30 | 1.25±1.27 | 0.85±0.95 | |
| Spark Ignition | 2.14±1.65 | 1.11±0.91 | 2.55±2.09 | 2.57±1.93 | 3.01±2.40 |
| Highway vehicle | | | | 1.30±1.26 | |
| Diesel | 0.43±0.34 | 0.45±0.34 | | 2.16±1.32 | 0.35±0.36 |

RESULTS AND DISCUSSION

The average source contributions are summarized in Table 1. Similar to previous studies in the eastern US, secondary sulfate is the most important source. On average, it contributes 38-51% of concentration for PM_{2.5} mass at these sites. Given the concentrations observed at Chester, these analyses suggest that more than 93% of sulfate measured in the New York metropolitan area are transported from distance sources. Secondary nitrate is a major component of PM_{2.5}. It contributes about 8-18% of concentration for PM_{2.5}. About 54 to 65% of the ammonium nitrate measured in metropolitan area of New York City is the result of transport based on the Chester concentrations. The contributions of soil dust to PM_{2.5} are 6-11% at five STN sites. According to EPA's National Emission Inventories, fugitive dust is the most important primary PM_{2.5} emission source in this area. It accounts about 38-57% of local emissions of PM_{2.5} in the Bronx, Queens, Union and Morris counties (Table 2). Highway vehicle emissions account for about 6 to 12% local emission. The oil combustion contributes about 4-11% of concentration for PM_{2.5} at these four sites. It is much higher than the EPA estimated contribution of oil burning to primary emission sources of PM_{2.5} listed in Table 2. The source contributions of spark ignition vehicles are 8 to 22% at the five sites. They are higher than EPA highway vehicle contribution to primary emission sources of PM_{2.5} listed in Table 2. Diesel emissions contribute between 3 and 15% of the PM_{2.5} at the five sites. Aged sea salt contributes about 3 to 7% of concentration for PM_{2.5} at the five sites. Wood burning is listed by EPA as an import primary emission source for PM_{2.5} in Queens, Union and Morris counties. Wood burning is supposed to account 16.7, 17.3 and 19.7% of primary PM_{2.5} emission in these three counties. However, there does not seem to be sufficient measured potassium concentrations that such emissions would be expected to generate (Watson *et al.*, 2001) and thus, wood burning could be identified at any of the sites.