

Short-Term Temporal Variation in PM_{2.5} Mass and Chemical Composition during the Atlanta Supersite Experiment, 1999

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ABSTRACT

Measurements in urban Atlanta of transient aerosol events in which PM_{2.5} mass concentrations rapidly rise and fall over a period of 3–6 hr are reported. The data are based on new measurement techniques demonstrated at the U.S. Environmental Protection Agency (EPA) Atlanta Supersite Experiment in August 1999. These independent instruments for aerosol chemical speciation of NO₃⁻, SO₄²⁻, NH₄⁺, and organic and elemental carbon (OC and EC), reconstructed the observed hourly dry PM_{2.5} mass to within 20% or better. Data from the experiment indicated that transient PM_{2.5} events were ubiquitous in Atlanta and were typically characterized by a sudden increase of EC (soot) and OC in the early morning or SO₄²⁻ in the late afternoon. The frequent temporal decoupling of these events provides insights into their origins, suggesting mobile sources in metro Atlanta as the main contributor to early morning PM_{2.5} and more regionally located point SO₂ sources for afternoon PM_{2.5} events. The transient events may also have health implications. New data suggest that short-term PM_{2.5} exposures may lead to adverse health effects.¹ Standard integrated filter-based

techniques used in PM_{2.5} compliance monitoring networks and in most past PM_{2.5} epidemiologic studies collect samples over 24-hr periods and thus are unable to capture these transient events. Moreover, health-effects studies that focus on daily PM_{2.5} mass alone cannot evaluate the health implications of the unique and variable chemical properties of these episodes.

INTRODUCTION

PM_{2.5} includes all particles with aerodynamic diameters less than 2.5 μm. These particles scatter and absorb light, interact with and affect clouds, and deposit within human respiratory systems. Their presence in the atmosphere has been linked to the formation of haze and changes in the atmosphere's radiative balance,² as well as adverse effects on human health, crops, and materials.³ In the case of human health effects, epidemiologic studies suggest a connection between increases in the mass concentration of PM_{2.5} (typically expressed in units of μg/m³) and increased morbidity and mortality through pulmonary and cardiovascular diseases in susceptible populations.⁴⁻⁷ The identification of the role of various PM constituents in PM toxicity is an area of active current research. One major focus has been on combustion emissions, such as elemental carbon (EC; soot) and organic carbon (OC) from mobile sources and sulfates from coal-fired power generators.⁸

For the most part, the association between PM_{2.5} and human health has been established through epidemiologic studies in which ambient aerosol concentrations were monitored by collecting particles on filters over extended time periods (e.g., 24 hr) and then using standard analytical techniques to determine the total mass and composition of the collected particles. The U.S. Environmental Protection Agency (EPA) has promulgated two National Ambient Air Quality Standards for PM_{2.5}, one based on a max 24-hr averaged concentration of 65 μg/m³ and the other on an annual average of 15 μg/m³.⁹ However, shorter-term, acute exposures caused by a rapid increase in PM_{2.5} concentrations or an increase in one chemical component may also have adverse health

IMPLICATIONS

Newly developed instruments for continuous highly time-resolved measurements of fine particle mass and chemical composition were deployed in metro Atlanta for the EPA Atlanta Supersite Experiment, August 1999. Measurements revealed the ubiquitous presence of transient PM_{2.5} episodes in which particle mass rapidly rises and falls over a period of a few hours but which go undetected with traditional time-integrated measurements. Many of these transient events result in an increase in PM_{2.5} mass by factors of 2–3. Speciated composition data show that these events are driven by sudden increases of two specific aerosol chemical components that dominate at different times, carbonaceous events in the early morning and SO₄²⁻ events in late afternoon. Apart from providing insights into sources, the unique chemical nature of these transient events may have specific health effects that previous epidemiologic studies based on highly averaged aerosol data could not readily resolve.

effects. Historically, characterization of the effects of such transient exposures has been problematic because of measurement limitations. This situation is changing as a result of the development of near-real-time aerosol monitors. For example, a recent study using high-resolution measurements of $PM_{2.5}$ mass suggests that short-term spikes in $PM_{2.5}$ mass can trigger the onset of myocardial infarction within a few hours of a pollution event.¹ The effective mitigation of fine particle pollution and its harmful effects on human health and the environment requires an understanding of the sources and properties of these particles as well as the processes that determine these properties. This work attempts to elucidate some of these key aspects of $PM_{2.5}$ in a relatively polluted urban environment by analyzing the near-real-time measurements of $PM_{2.5}$ mass concentration and chemical composition gathered during the Atlanta Supersite Experiment.

EXPERIMENTAL METHODS

The Atlanta Supersite Experiment was conducted from August 3, 1999, to September 1, 1999, at a ground-based site located in a mixed residential and industrial neighborhood approximately 4 km northwest of downtown Atlanta. An overview of the experiment is provided by Solomon et al.¹⁰ As part of this experiment, a wide range of instrumentation for both aerosol and gas-phase measurements was deployed. This included a variety of instruments capable of near-real-time quantification of $PM_{2.5}$ mass and chemical composition with a time resolution of 1 hr or less. Among the $PM_{2.5}$ chemical components measured by these so-called semicontinuous instruments were SO_4^{2-} , NO_3^- , NH_4^+ , OC, and EC (soot, or black carbon). Some of the instruments used were designed to measure only one $PM_{2.5}$ component (e.g., SO_4^{2-}), others were designed to measure multiple components (e.g., OC and EC, or numerous aerosol ions). Results from intercomparisons of the various semicontinuous instruments for the measurement of NO_3^- and SO_4^{2-} , and OC and EC are reported elsewhere.^{11,12} The data used for this paper are based on averaging various semicontinuous measurements of the same species. The acronyms used to identify the institutions operating the instruments are defined at the end of this paper and are those used in the intercomparison papers where a more complete description of the instrumentation is provided.^{11,12} The NH_4^+ reported here is, for example, the average of the two semicontinuous NH_4^+ techniques deployed during the experiment, both based on ion chromatograph (IC) analysis^{13,14} (operated by ECN and GT/BL, respectively). SO_4^{2-} and NO_3^- are the average of three IC-based detectors (ECN, GT/BL, and TT) and a flash vaporization

approach¹⁵ (ADI). Reported OC is the average of an in situ thermal-optical carbon analyzer¹⁶ (RU/OGI), the Rupprecht and Patashnick (R&P) 5400 ambient carbon analyzer, and a flash vaporization carbon analyzer (ADI). In addition, the RU/OGI and the R&P 5400 measured EC, which was also averaged with EC inferred from a Radiance Research particle soot absorption photometer (PSAP, operated by ARA) and the Magee Scientific AE-16 aethalometer (operated by HSPH).

Near-real-time dry $PM_{2.5}$ mass was measured during the Atlanta experiment using a tempered element oscillatory microbalance (TEOM) (R&P) with the sensor heated to 50 °C to drive off condensed water.¹⁷ The TEOM measures the mass collected on a small filter oscillating at the end of a hollow glass tube through which sample air is drawn. Changes in the natural frequency of this oscillation are related to changes in filter mass. Possible aerosol volatility artifacts associated with running the TEOM at the high temperature of 50 °C are discussed in the following section. Our analysis of data from these instruments indicate that transient $PM_{2.5}$ events are frequent in Atlanta and have specific chemical signatures that provide clues to their sources and may have specific health consequences.

RESULTS AND DISCUSSION

Mass Closure

Before analyzing the data from the near-real-time instrumentation for insights into the characteristics of $PM_{2.5}$ during the Atlanta experiment, the completeness and precision of the chemical speciation is assessed by testing for mass closure. This is done by comparing the sum of the measured chemical components to an independent gravimetric measurement of the total dry mass. Because OC measurements only quantify the carbon component of OC, mass closure requires a correction factor to convert mass of carbon to mass of organic compounds.

$$OM = OC \times (\text{correction factor}) \quad (1)$$

where OC is the measured mass of organic C in $PM_{2.5}$ and OM is the estimated total organic mass of the organic compounds (including O, N, etc.) in $PM_{2.5}$. The correction factor is the average organic molecular weight per carbon weight. In keeping with many previous investigators, a correction factor of 1.4 is assumed. Values as high as 1.6 ± 0.2 for urban aerosols are reasonable.¹⁸ Heating the TEOM sensor (i.e., filter) to obtain a dry $PM_{2.5}$ mass measurement can result in an underestimate of $PM_{2.5}$ mass caused by loss of volatile aerosol components and may offset to some extent the use of a low correction factor for OC.

In general, the agreement between the reconstructed mass, using the measurements for the $PM_{2.5}$ constituents and the directly measured $PM_{2.5}$ is quite good. For the entire study period, the mean $PM_{2.5}$ mass and standard deviation obtained from summing the chemical components was $29.5 \pm 10.8 \mu\text{g}/\text{m}^3$, while the mean and standard deviation obtained with the TEOM was $30.9 \pm 11.7 \mu\text{g}/\text{m}^3$. The average mass obtained from the TEOM itself was about 4% lower than that obtained from the integrated filter samplers used during the study.¹⁹

Higher resolution measurements can more rigorously assess mass closure. For the one-month study, Figure 1 shows the average diurnal $PM_{2.5}$ concentration and its major chemical constituents. Comparison of the sum of the major constituents to the measured $PM_{2.5}$ mass reveals generally good agreement. The largest discrepancies are found at 0900 EST when the reconstructed mass exceeds the measured mass by about 10%, and after 1400 EST when the measured mass exceeds the reconstructed mass by about 7–12%. Nonetheless, a similar 24-hr trend is seen in both sets of measurements.

Mass reconstruction based on the 1-hr data for the one-month study is shown in Figure 2. This figure shows that even when the comparison is made using the hourly-averaged data the agreement is generally 20% or better, although exceptions having discrepancies of 40% or more occurred on occasion. Pearson product linear regression of the sum of OM, EC, NO_3^- , SO_4^{2-} , and NH_4^+ versus $PM_{2.5}$ in $\mu\text{g}/\text{m}^3$ is

$$\begin{aligned} \text{Speciated Sum} &= 2.78 \pm 0.93 + 0.89 \pm 0.03 \times PM_{2.5}, \\ r^2 &= 0.879 \quad (2) \end{aligned}$$

where the uncertainties are based on 95% confidence intervals. Although this is the first chemical mass reconstruction at 1-hr time resolution over an extended period of time, the agreement may be somewhat fortuitous because of compensating errors between the speciated sum and the mass measurement. At the very least, the 1-hr reconstruction shown in Figure 2 demonstrates consistency between these diverse instruments and their ability to document the occurrence and chemical characteristics of short-term, transient $PM_{2.5}$ mass variations.

Transient Characteristics of Atlanta $PM_{2.5}$ Aerosol during the Supersite Experiment

In Atlanta, based on measurements extending over a one-year period, total carbon (EC+OM), and SO_4^{2-} each typically comprises about 35–45% of the $PM_{2.5}$ dry mass, with the remainder being composed primarily of NH_4^+ and NO_3^- .²⁰ The daily averages for the one-month Supersite Experiment, shown in Figure 1, are consistent with these more general yearly trends.

Figure 1 indicates that, on average, $PM_{2.5}$ exhibited relatively modest variations (of 20% or less) over a diurnal cycle during the Atlanta experiment. In contrast, the hourly averaged data from the semicontinuous monitors, illustrated in Figures 3b and 4b, indicate that the variations on short time scales were quite large. In several instances, $PM_{2.5}$ was observed to vary by factors of 2–3 over time intervals of a few hours. Because SO_4^{2-} and EC+OM are the two major components of $PM_{2.5}$, the short-term variations in $PM_{2.5}$ illustrated in Figures 3b and 4b are most likely associated with variations in SO_4^{2-} and EC+OM. Interestingly, however, the results illustrated in Figure 1 suggest that, at least on average, SO_4^{2-} and EC+OM appear to vary independently of each other, with SO_4^{2-} tending to peak in the late afternoon and EC+OM peaking in the morning. In the analyses presented here, these tendencies are examined in more detail.

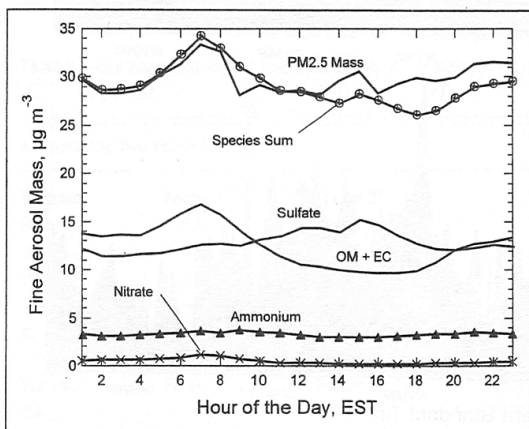


Figure 1. Atlanta Supersite experiment average concentrations for August 4–31, 1999, based on hourly averaged data from the semicontinuous detectors. Organic matter (OM) is the measured OC multiplied by 1.4.

Multivariate Analysis. An objective assessment of the contributions of SO_4^{2-} and EC+OM to variations in $PM_{2.5}$ can be obtained through a multivariate analysis in which a linear regression is used to simultaneously apportion the variances in $PM_{2.5}$, SO_4^{2-} , and EC+OM to a number of underlying factors.²¹ The relative importance of each factor to each variable is then indicated by the so-called loading derived for that variable and factor. A positive (negative) loading indicates that

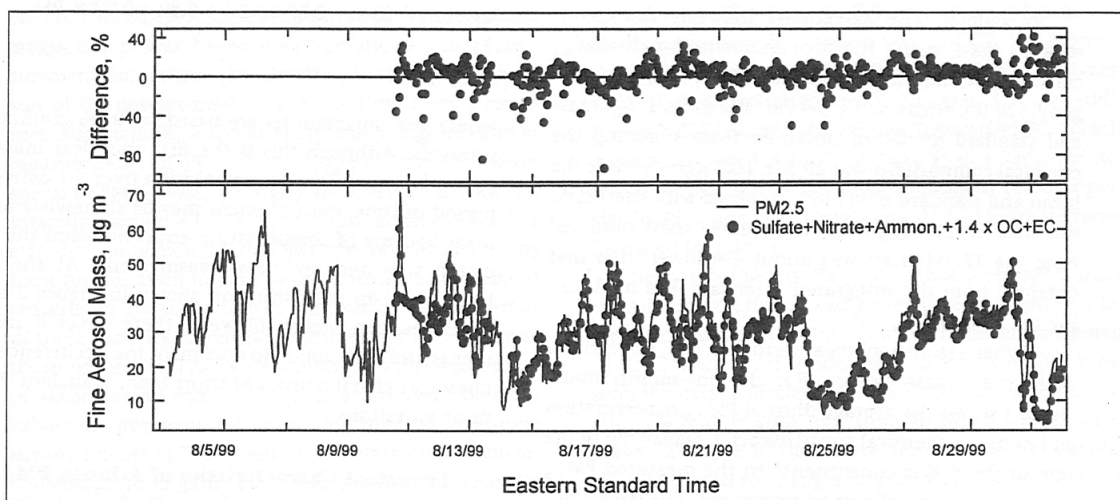


Figure 2. $PM_{2.5}$ mass reconstruction from Atlanta Supersite measurements. Dry $PM_{2.5}$ (TEOM sensor heated to 50 °C) is compared with the sum of the SO_4^{2-} , NO_3^- , NH_4^+ , OC times 1.4, and EC. The top plot shows the percent difference between speciated sum and mass. Data are 1-hr averages.

the variable is positively (negatively) correlated to that factor, and a small loading indicates that the variable is not affected by that factor. The square of a loading for

any given factor and variable is the fraction of the total variance in that variable that can be explained by that factor. Table 1 summarizes the results of a multivariate

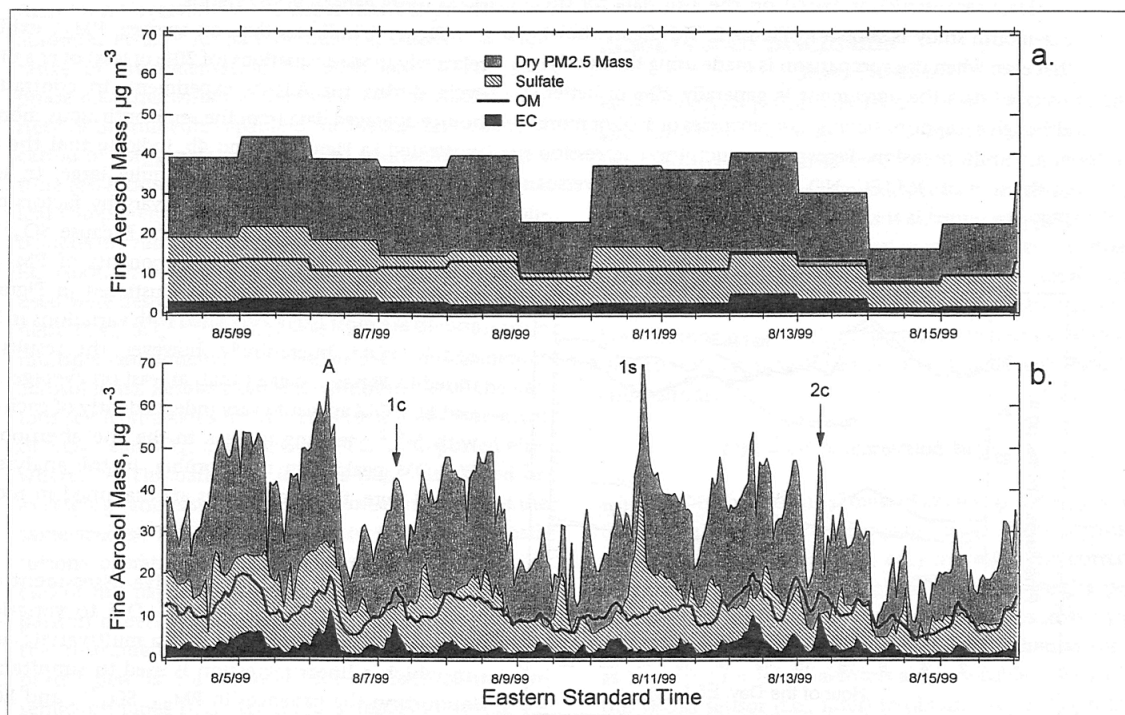


Figure 3. Chemical composition of the fine aerosol compared with the total $PM_{2.5}$ mass for the first half of the Supersite experiment. Plot (a) shows results from a 24-hr average and (b) 1-hr average. In plot (b) of Figure 3 and 4, specific events in which the peak in $PM_{2.5}$ is driven by SO_4^{2-} (1s–4s) and by EC+OM (1c–4c) are shown in the graph. Table 1 summarizes pertinent measurements at these times. Note that in some cases, the EC+OM events are also associated with SO_4^{2-} ; examples are labeled with letters A–C.

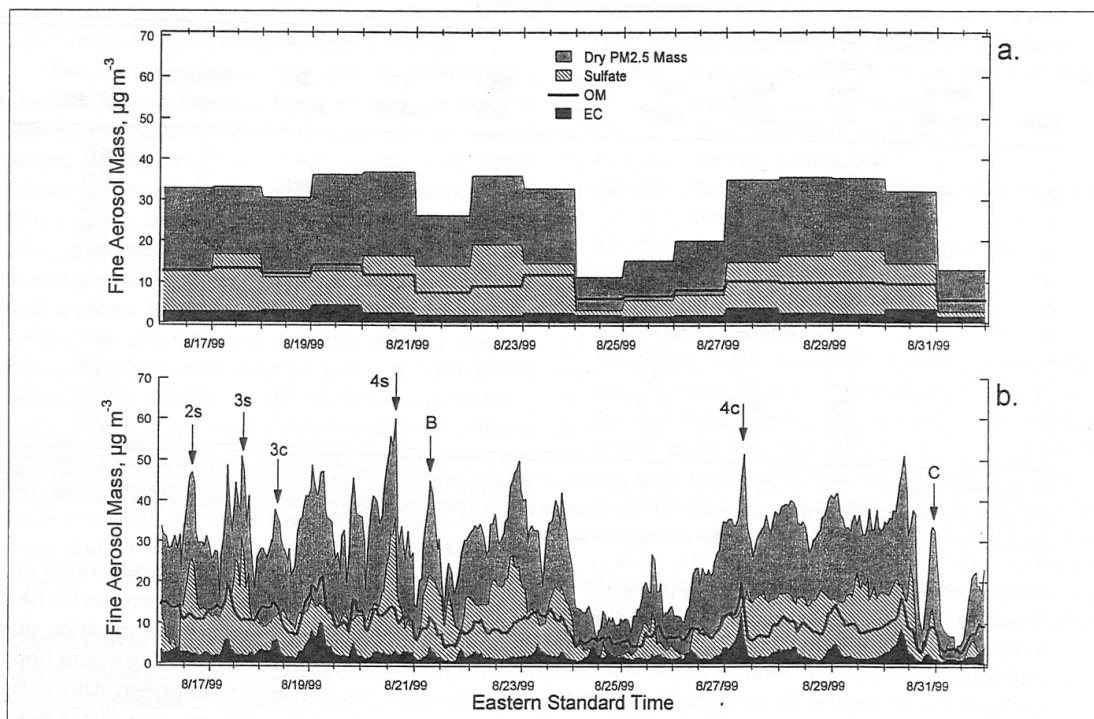


Figure 4. Same as Figure 3, except for the second half of the Supersite experiment.

analysis involving the 1-hr averaged values observed for $PM_{2.5}$, SO_4^{2-} , and $EC+OM$. This analysis shows that about 97% of the total variance in $PM_{2.5}$ mass can be explained by two independent factors; one being most strongly associated with SO_4^{2-} and the other with $EC+OM$.

Table 1. Factor loadings derived from multivariate analysis of 1-hr averaged $PM_{2.5}$, SO_4^{2-} , and $EC+OM$.

A. Assuming Two Factors			
Variable	Factor 1 ^a	Factor 2 ^a	
$PM_{2.5}$	0.90	0.38	
SO_4^{2-}	0.99	—	
$EC+OM$	0.43	0.90	
B. Assuming Three Factors			
Variable	Factor 1 ^a	Factor 2 ^a	Factor 3 ^a
$PM_{2.5}$	0.89	0.37	0.25
SO_4^{2-}	0.98	—	—
$EC+OM$	0.41	0.91	—

^aFactor loadings that explain less than 5% of the variance in any variable were deemed to not be significant and have been replaced by "—."

Transient Events. A feature revealed by a higher time-resolution measurement is the presence of short-term transient events. A comparison in Figures 3 and 4 of the variation in $PM_{2.5}$ mass and its chemical components as a function of time using 1-hr and 24-hr averages demonstrates this. It is apparent that, on a time scale of hours, $PM_{2.5}$ was highly variable and that the use of 24-hr averages largely masks this variability. For example, on August 10, the 1-hr averaged $PM_{2.5}$ dry mass reached $70.7 \mu\text{g}/\text{m}^3$, the highest value attained during the study and almost double the 24-hr average mass for the day of $37.2 \mu\text{g}/\text{m}^3$ (this episode is labeled as 1s in Figure 3b and Table 2). From the onset of the event to the time of peak concentration (roughly 4 hr), the 1-hr average $PM_{2.5}$ increased from 29.0 to $70.7 \mu\text{g}/\text{m}^3$. A recent study linking transient $PM_{2.5}$ events to health effects suggests that a rapid change in $PM_{2.5}$ mass may be as or more important to health than the peak concentration reached during the episode.¹ Inspection of Figures 3 and 4 reveals that the Atlanta data are replete with similar, albeit less intense, short-term events that appear in the 1-hr but not in the 24-hr averages.

The timing and chemical nature of the transient $PM_{2.5}$ events provide clues to their origin. In Atlanta, the aerosol chemical speciation data and our multivariate

Table 2. Pertinent parameters recorded during transient episodes identified in Figures 3 and 4.

Label	Day of Aug 1999	Hour at		PM _{2.5} μg/m ³	SO ₄ ²⁻ μg/m ³	NO ₃ ⁻ μg/m ³	NH ₄ ⁺ μg/m ³	OC μgC/m ³	EC μgC/m ³	Acidity ^a neq/m ³	SO ₄ ²⁻ /PM	TC/PM ^b
		Peak EST										
1c	7	6:30	43.1	11.8	0.9	NA	11.8	7.8	NA	0.27	0.56	
2c	12	8:30	53.9	14.3	1.3	4.0	15.0	10.9	99	0.27	0.59	
3c	18	7:30	37.8	11.4	0.6	3.0	10.7	5.5	79	0.30	0.54	
4c	27	7:30	48.6	14.2	1.9	4.8	12.2	9.4	65	0.29	0.54	
1s ^c	10	17:00	60.2	37.7	0.2	6.8	8.1	2.1	439	0.63	0.22	
2s	16	15:30	46.8	25.6	0.6	5.1	8.3	2.5	262	0.55	0.30	
3s	17	15:30	50.8	30.8	0.3	4.0	7.6	2.0	421	0.61	0.25	
4s	20	15:30	60.1	35.8	0.4	5.2	9.9	3.1	465	0.60	0.28	
A	6	6:30	65.7	27.6	0.9	NA	12.2	8.7	NA	0.42	0.39	
B	21	7:30	45.0	21.9	3.1	7.9	8.1	3.9	67	0.49	0.34	
C	30	22:30	33.8	13.6	0.2	4.0	7.1	1.7	63	0.40	0.34	

^aThe apparent acidity equals $2 \times \text{SO}_4^{2-} + \text{NO}_3^- - \text{NH}_4^+$ with all concentrations expressed in nmole/m³; ^bTC/PM is the total carbon ($\text{OC} \times 1.4 + \text{EC}$) divided by fine aerosol mass (PM_{2.5}); ^cBecause the sulfate peaked 1 hr prior to the PM_{2.5} in this case, the average over 2 hr is given here. At 1630 hr the sulfate peak was 41.2 μg/m³ and the PM_{2.5} 49.8 μg/m³, in the following hour, 1730, the sulfate was 34.2 μg/m³ and PM_{2.5} 70.7 μg/m³.

analysis show that, in most cases, the variability is driven by variations in either OC+EC or in SO₄²⁻, and as shown by the daily averages (Figure 1), as well as the multivariate analysis, they tend to vary independently. For example, in Figures 3b and 4b, four episodes in which OM (recall, OM = 1.4 × OC) and EC drive the PM_{2.5} mass peak are identified as 1c–4c. Likewise, four events when the PM_{2.5} peak is driven by a SO₄²⁻ increase are identified as 1s–4s. Three events when the peak PM_{2.5} was driven by simultaneous increases in all three components are labeled as A–C. Pertinent parameters associated with these events are summarized in Table 2. Combined, Table 2 and Figures 3 and 4 show that the OM+EC events often begin to develop in the early morning and peak between 0600 and 0800 EST. Transient OM+EC events were rarely observed in the afternoon. In contrast, most SO₄²⁻ events typically occur in mid- to late afternoon (1500–1700 EST); however, a few events did occur in the early morning at the time of the OM+EC events.

It is generally believed that these EC peaks with accompanying OC are from primary pollutants emitted directly into the atmosphere,²² while SO₄²⁻ is a secondary pollutant generated photochemically from the oxidation of SO₂.²³ Daily peaks will also depend on meteorological considerations. For example, after sunset, surface radiative cooling causes increased atmospheric stability and development of a low-level inversion. This tends to limit dispersion of surface-emitted pollutants and causes their near-surface concentrations to accumulate (e.g., EC and primary OC). After sunrise, however, surface heating promotes turbulence and downward mixing of air from above. This expands the surface

layer and dilutes near-surface concentrations of locally emitted pollutants. However, it increases the likelihood of spikes in the near-surface concentration of pollutants from more distant emissions by mixing upper-level air down to the surface. Consistent daily patterns in wind speed and direction may also play a role in producing the events. These trends are generally consistent with the following hypothesis: the transient EC and OM (OC) events are from urban mobile sources and SO₄²⁻ is from the more distant tall stacks of coal-fired power plants that surround the Atlanta metropolitan area.

Implications of Transient Events, Aerosol Acidity, and Possible Health Effects

During the carbonaceous events, the fraction of total carbon (EC+OM) to PM_{2.5} varied between 54 and 59%, compared with the study average of 43%. For the SO₄²⁻ events, the SO₄²⁻ mass fractions (SO₄²⁻/PM_{2.5}) varied between 55 and 63%, and the study average was 44%. A consequence of the high SO₄²⁻ concentrations during these events is typically a 4-fold increase in the PM_{2.5} apparent acidity, as shown in Table 2. Here, apparent acidity is the difference in the measured concentrations of anions and cations, all expressed in equivalents. Because the inorganic composition of the PM_{2.5} aerosol during the study was dominated by SO₄²⁻ and NH₄⁺ (see Figure 1), the apparent acidity is well represented by $2 \times \text{SO}_4^{2-} - \text{NH}_4^+$. Alternatively, the molar ratio of NH₄⁺ to SO₄²⁻ can be calculated from the measured concentrations. For the 1-month study, the average NH₄⁺/SO₄²⁻ molar ratio was 1.7, close to neutral (NH₄)₂SO₄. The four episodes

identified as 1 s, 2 s, 3 s, and 4 s in Figures 3b and 4b had NH_4^+ -to- SO_4^{2-} molar ratios between 1 and 0.7.

The differing diurnal patterns of specific $\text{PM}_{2.5}$ chemical components may be important from a health-effects and epidemiologic study point of view. For example, the late-afternoon SO_4^{2-} events occur when people tend to be active and results in exposure to high $\text{PM}_{2.5}$ acidity that accompanies these episodes.⁷ Moreover, there may be synergistic interactions with ozone that also typically peaks in the late afternoon in Atlanta and in other urban areas. In terms of epidemiologic studies, the differing diurnal trends of $\text{PM}_{2.5}$ carbonaceous and SO_4^{2-} mass could be used in a health-effects study similar to Peters et al.¹ to relate transient $\text{PM}_{2.5}$ health effects to specific aerosol chemical components. Previous epidemiologic studies were ill suited to identify these effects. This is because 24-hr filter sampling times would fail to resolve these transient episodes and their diurnal trends, and the studies were based on reported daily health statistics.

CONCLUSION

Newly developed instrumentation for high time resolution measurements of aerosol chemical composition and mass deployed at the EPA Atlanta Supersite show that transient events frequently occur in which $\text{PM}_{2.5}$ mass concentrations rapidly rise and fall over a period of 3–6 hr. The contributions of these components to PM-induced toxicity is poorly understood and a scientific issue of much current interest. In Atlanta, these events are driven primarily by peaks in the carbonaceous or SO_4^{2-} components of $\text{PM}_{2.5}$. These components have different sources and generally impact the $\text{PM}_{2.5}$ mass concentration at different times of the day. A recent epidemiologic study suggests transient $\text{PM}_{2.5}$ events can have adverse health effects;¹ however, current $\text{PM}_{2.5}$ monitoring networks using 24-hr averaged filter samples are unable to resolve such events.

The spatial extent of the EC+OC and SO_4^{2-} events described here and their ultimate impact on the magnitude and character of the $\text{PM}_{2.5}$ exposure of citizens living in metropolitan areas such as Atlanta cannot be determined from this study alone. However, future studies involving the deployment of real-time chemical speciation monitors at a number of sites throughout the metropolitan area could better define the scale of these events. Combined with meteorological measurements, this network could help in identifying specific sources responsible for the transient events. The measurements could also be combined with relevant real-time epidemiologic data to assess possible health impacts on the metropolitan population.

ACRONYMS

ECN:	Netherlands Energy Research Foundation
GT/BL:	Georgia Institute of Technology and Brookhaven National Laboratory
TT:	Texas Institute of Technology
ADI:	Aerosol Dynamics Inc.
RU/OGI:	Rutgers University and Oregon Graduate Institute
ARA:	Atmospheric Research and Analysis
HSPH:	Harvard School of Public Health

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DISCLAIMER

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