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Measurement, Analysis, and Modeling of Fine Particulate Matter in Eastern North Carolina

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ABSTRACT

An analysis of fine particulate data in eastern North Carolina was conducted to investigate the impact of the hog industry and its emissions of ammonia into the atmosphere. The fine particulate data are simulated using ISORROPIA, an equilibrium thermodynamic model that simulates the gas and aerosol equilibrium of inorganic atmospheric species. The observational data analyses show that the major constituents of fine particulate matter ($PM_{2.5}$) are organic carbon, elemental carbon, sulfate, nitrate, and ammonium. The observed $PM_{2.5}$ concentration is positively correlated with temperature but anticorrelated with wind speed. The correlation between $PM_{2.5}$ and wind direction at some locations suggests an impact of ammonia emissions from hog facilities on $PM_{2.5}$ formation. The modeled results are in good agreement with observations, with slightly better agreement at urban sites than at rural sites. The predicted total inorganic particulate matter (PM) concentrations are within 5% of the observed values under conditions with median initial total PM species concentrations, median relative humidity (RH), and median temperature. Ambient conditions with high PM precursor concentrations, low temperature, and high RH appear to favor the formation of secondary PM.

INTRODUCTION

Particulate matter (PM) has become a relatively recent concern in the overall air quality of our environment. In 1997, the U.S. Environmental Protection Agency (EPA) modified the National Ambient Air Quality Standards for PM by dividing the total suspended particulate standard into two separate modes of particulates, fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) particles, with the standards for $PM_{2.5}$ being 65 $\mu g \cdot m^{-3}$ daily and 15 $\mu g \cdot m^{-3}$ annually. EPA has recently tightened the daily average standard for $PM_{2.5}$ to be 35 $\mu g \cdot m^{-3}$. $PM_{2.5}$ is known to contribute to human respiratory problems, dry and wet acidic deposition, reduced visibility, and radiative forcing.[1] $PM_{2.5}$ is composed of primary and secondary pollutants; primary $PM_{2.5}$ species may include organic carbon (OC), elemental carbon (EC), soil dust, ash, and sulfate. Secondary $PM_{2.5}$ may include sulfate, nitrate, ammonium, and OC, which are formed through the oxidation of their gas-phase precursors such as sulfur dioxide, nitrogen dioxide, ammonia (NH_3), and volatile organic compounds (VOCs).

IMPLICATIONS

The role of gaseous ammonia in $PM_{2.5}$ mass and composition concentration was determined. These emissions of ammonia from hog confined animal feeding operations are responsible for the increase in $PM_{2.5}$ concentrations, elucidating the role of ammonia in rural air quality.

In particular areas of the United States, NH_3 and ammonium have become significant contributors to total $PM_{2.5}$ concentration. NH_3 can react with acidic compounds to form various aerosols such as ammonium nitrate (NH_4NO_3), ammonium chloride (NH_4Cl), ammonium sulfate ($(NH_4)_2SO_4$), and ammonium bisulfate (NH_4HSO_4). Globally, it is estimated that a total of 49.3 Tg of NH_3 is emitted into the atmosphere, with 56% of this total being anthropogenic. The largest contributor to these NH_3 emissions is domestic animal waste decomposition, which accounts for 22 Tg NH_3. The other major continental sources of NH_3 emissions include soil loss from organic matter, fertilizer release, biomass burning, and coal combustion, which account for 10, 4, 1.3, and 0.03 Tg/yr respectively.[2–5] In the state of North Carolina alone, the largest source of NH_3 emission is domestic animal waste.[6]

In recent years, the hog industry of North Carolina has experienced rapid growth. Between 1986 and 2005, the hog population expanded from 2.4 to 9.7 million, which makes it rank second in terms of pig production by state nationwide.[7] The swine in North Carolina are estimated to emit 68,540 t NH_3/yr, which makes swine the largest contributor among all domesticated animals in North Carolina.[8] These swine are concentrated in the coastal plain region of the state or the southeast corner covering Bladen, Duplin, Greene, Lenoir, Sampson, and Wayne counties.[9] Promising results have been reported for reducing NH_3 from swine manure through the use of an "engineered system," that is, a treatment plant with solid-liquid separation. Szogi[10] reported a 73% reduction in NH_3 emissions from the implementation of such a system.

Several aerosol modules have been developed to simulate $PM_{2.5}$. A particular area of focus has been studying the inorganic aerosols of $PM_{2.5}$, which make up 25–50% of total $PM_{2.5}$.[11] Some examples of these aerosol modules are MARS-A, SEQULIB, SCAPE2, EQUISOLV II, and AIM2, which have been thoroughly reviewed for their similarities and differences.[12] ISORROPIA is a thermodynamic equilibrium model used for predicting the partitioning of major inorganic species between the gas phase and aerosol phase. This model was selected because of its efficiency in computation and its overall satisfactory performance

Table 1. Specifications of measured $PM_{2.5}$ data at the sampling sites in North Carolina.

Site Names	Time Period of Sampling	Number of Points	Site Type	Kind of Sample
Fayetteville	January 2002 to January 2004	124	Urban	Speciated $PM_{2.5}$ concentrations
Goldsboro	January 2001 to December 2003	362	Rural	$PM_{2.5}$ concentrations
Jacksonville	January 2001 to December 2003	354	Coastal	$PM_{2.5}$ concentrations
Kenansville	January 2001 to December 2003	361	Rural	$PM_{2.5}$ concentrations
Kinston	January 2002 to January 2004	123	Rural	Speciated $PM_{2.5}$ concentrations
Kinston	January 2001 to December 2003	360	Rural	$PM_{2.5}$ concentrations
Raleigh	January 2002 to January 2004	146	Urban	Speciated $PM_{2.5}$ concentrations
Raleigh	January 2001 to December 2003	1084	Urban	$PM_{2.5}$ concentrations
Wilmington	January 2001 to December 2003	348	Coastal	$PM_{2.5}$ concentrations

against more comprehensive aerosol thermodynamic models. With an input of temperature, relative humidity (RH), and the total (gas + aerosol) concentrations of sodium, ammonium, nitrate, chloride, and sulfate, ISORROPIA predicts how much the total amount will be in the gas and aerosol phases.[13,14]

The primary objective of this study was to investigate the effect of increased NH_3 emissions on the $PM_{2.5}$ concentrations on the surrounding cities in North Carolina. The source of these increased NH_3 emissions is the presence of the hog industry. The work conducted here includes analysis of the constituents of $PM_{2.5}$, their correlations with meteorological variables, and the impact of the hog facilities on $PM_{2.5}$ concentrations. Another objective was to test how well ISORROPIA can predict the $PM_{2.5}$ concentrations and under what ambient conditions the model has its best performance in reproducing $PM_{2.5}$ concentrations.

MEASUREMENT AND MODELING METHODS

$PM_{2.5}$ observational data were obtained from the North Carolina Division of Air Quality (http://daq.state.nc.us/). These data consist of average daily values for seven sites in eastern North Carolina between 2001 and early 2004. The exact specifications of the particulate data are listed in Table 1. Fayetteville and Raleigh are the urban sites, which are situated to the west of the majority of the hog facilities. Goldsboro, Kenansville, and Kinston are the rural sites, with Kenansville being both the smallest city and the most enclosed by the hog facilities. Jacksonville and Wilmington are two coastal sites with the hog facilities to the north and west of their positions. Figure 1 shows the locations of the seven sites in North Carolina and their relative positions to hog facilities.[15] For all of these sites, when the average daily value consisted of less than 90% of the individual hours reporting, the average daily data point was considered inaccurate and was discarded. Meteorological data were obtained for each site from the North Carolina State Climate Office (http://www.nc-climate.ncsu.edu/). Although $PM_{2.5}$ data are available for all seven sites, speciated $PM_{2.5}$ data are only available for two urban sites, Fayetteville and Raleigh, and one rural site, Kinston.

The model was set for a forward problem, in which the total (both gas and aerosol) concentrations of ammonium, sulfate, sodium, chloride, and nitrate concentrations in addition to RH and temperature (T) were used to calculate the total aerosol mass. Also, the model was set to run in the thermodynamically stable state (i.e., solids can be formed when RH decreases below its deliquescence RH [DRH]) instead of the metastable state (i.e., aerosols are in liquid even when RH $<$ DRH). The initial conditions for

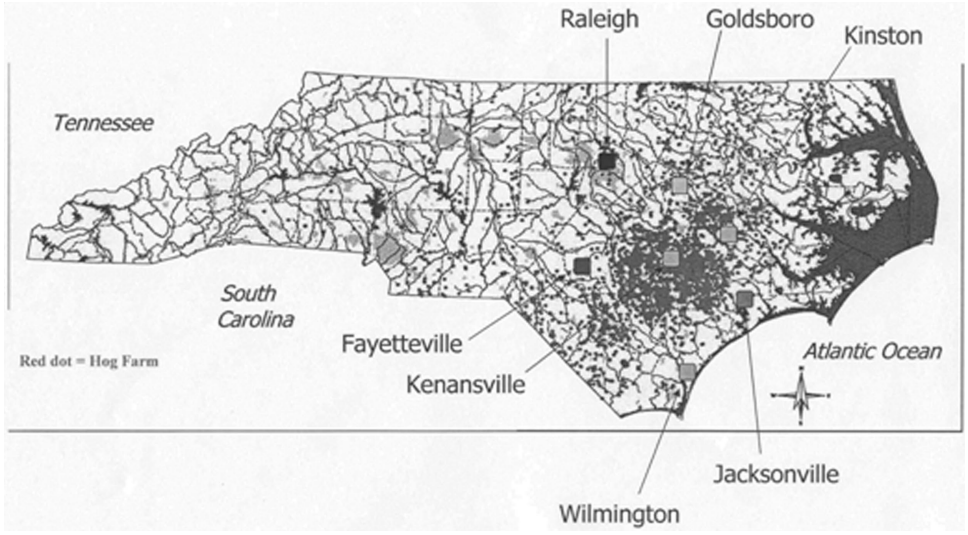

Figure 1. Map of hog facilities in North Carolina and $PM_{2.5}$ sampling sites.

the ISORROPIA model simulations are listed in Table 2. These conditions were selected based on available observational data in North Carolina and literature values when observational data were not available. For each modeled site (i.e., Kinston, Fayetteville, and Raleigh), three levels of initial total PM species were used: median, minimum, and maximum, representing the median, lower, and upper limits of the 2002 observations, respectively. For each concentration level, the model was run under three meteorological conditions: median RH/median T, minimum RH/maximum T, and maximum RH/minimum T. The output variables include concentrations of gaseous species (i.e., NH_3, hydrochloric acid, and nitric acid) and aerosol species (i.e., sulfate, ammonium, nitrate, sodium, chloride, and water), as well as the pH value.

OBSERVED $PM_{2.5}$ AND ITS CORRELATIONS WITH METEOROLOGICAL VARIABLES

The particulate data were first analyzed for their main constituents at the three sites with detailed speciated $PM_{2.5}$ data over the entirety of the sampling period, as shown in Figure 2. The plot shows the major constituents of $PM_{2.5}$ to be OC, sulfate, and ammonium, consistent with the results by Harrison et al.,[16] and Tanner et al.[17] The additional components of $PM_{2.5}$ include nitrate, EC, and over 50 trace elemental species. The $PM_{2.5}$ OC concentrations were higher in the urban areas because of large local emissions of primary OC and VOCs. The sulfate and ammonium emissions were found to be slightly larger in the rural site, which we believe to be due to the influence of the hog farming facilities in the rural area.

Figure 3 shows the scatter plots of $PM_{2.5}$ concentration versus RH at Raleigh, Kinston, and Wilmington that represent urban, rural, and coastal areas. High $PM_{2.5}$ concentrations (>20 $\mu g/m^3$) occurred with the range of RH between 60 and 90%, and this effect was more prominent in the urban areas. To account for the nonconstant variance shown in Figure 3, the data were transformed using a log function, and in each case the data exhibited the same trends that are shown in Figure 3. This trend supports the fact that the overall RH increases the film of water formed on the surface of the particles, favoring the formation of $PM_{2.5}$. Figure 4 shows the correlation between $PM_{2.5}$ concentrations and wind speeds at three sites. To account for the nonconstant variance shown in Figure 4, the data were transformed using a log function, and in each case the data exhibited the same trends that are shown in Figure 4. The observed anticorrelation between PM concentration and wind speed is consistent

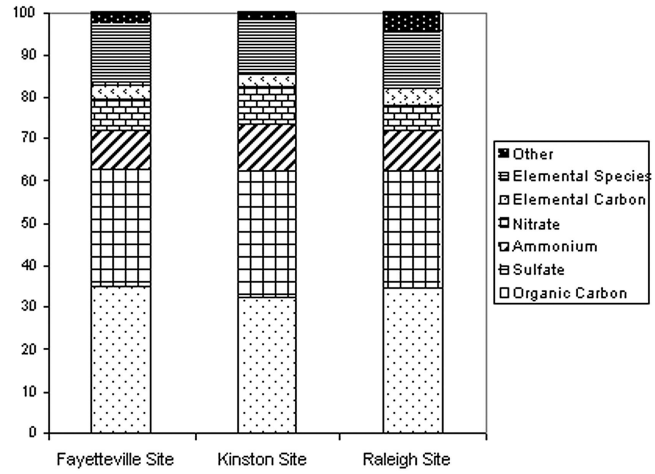

Figure 2. $PM_{2.5}$ composition at three speciated sites (Kinston, Fayetteville, and Raleigh) over entire sampling period.

with that of Chu et al.[18] and de Hartog et al.[19] The $PM_{2.5}$-T correlation plots for Raleigh, Kenansville, and Wilmington are shown in Figure 5 to represent urban, rural, and coastal areas, respectively. Many high $PM_{2.5}$ concentrations occurred at high T. The slopes range from 0.08 to 0.18 at the urban and the rural sites and 0.01 to 0.02 at the coastal site. To investigate the impact of NH_3 on $PM_{2.5}$ concentrations, the ammonium concentrations were plotted against the total $PM_{2.5}$ (figure not shown). The values for the slope, intercept, and the coefficient of determination are shown in Table 3. There are significant correlations in the two urban sites (i.e., Raleigh and Fayetteville), but no correlation at the rural site (i.e., Kinston). These values show that higher ammonium concentrations correlate with higher total $PM_{2.5}$ concentrations, which indicate a possible impact from the local NH_3 emissions on $PM_{2.5}$. The very low R^2 value in the Kinston correlation plot is thought to be due to the local variability of local primary OC $PM_{2.5}$ emissions (i.e., local biomass burning from farming practices). To investigate the correlation between wind direction and PM distributions, a box-whisker plot was made for all seven sites with respect to the eight cardinal directions, as shown in Figure 6. The minimum, 25th percentile, average, 75th percentile, and the maximum of each distribution are plotted. At each of the sites, the wind direction distribution was studied with the easterly and northerly winds as the highest contributors, but no single direction ever accounted for more than 25% of the total distribution. The impact of

Table 2. The initial species concentrations and meteorological conditions for ISORROPIA simulations.

Input Variables[a]		Kinston	Fayetteville	Raleigh
Sodium		0.22	0.19	0.17
Sulfate	Median, minimum, maximum	3.43, 0.58, 14.3	3.53, 0.75, 12.9	3.36, 0.72, 13.8
Ammonium	Median, minimum, maximum	3.13[b], 0.32, 11.5	3.19 [c], 0.07, 11.4	5.10[c], 0.83, 16.4
Nitrate	Median, minimum, maximum	1.07 [b], 0.26, 5.24	1.41[c], 0.18, 12.3	1.60[c], 0.19, 19.4
Chloride	Median, minimum, maximum	0.14 [b], 0.10, 0.97	0.33[c], 0.02, 3.20	0.34[c], 0.02, 4.75
RH (%)	Median, minimum, maximum	77, 46, 97	74, 38, 100	74, 36, 98
T (K)	Median, minimum, maximum	291.00, 269.61, 302.44	291.39, 271.22, 304.39	289.86, 268.72, 301.94

Notes: [a]All concentrations are given in $\mu g \cdot m^{-3}$; [b]Walker et al., 2004[21]; [c]Bari et al., 2003.[22]

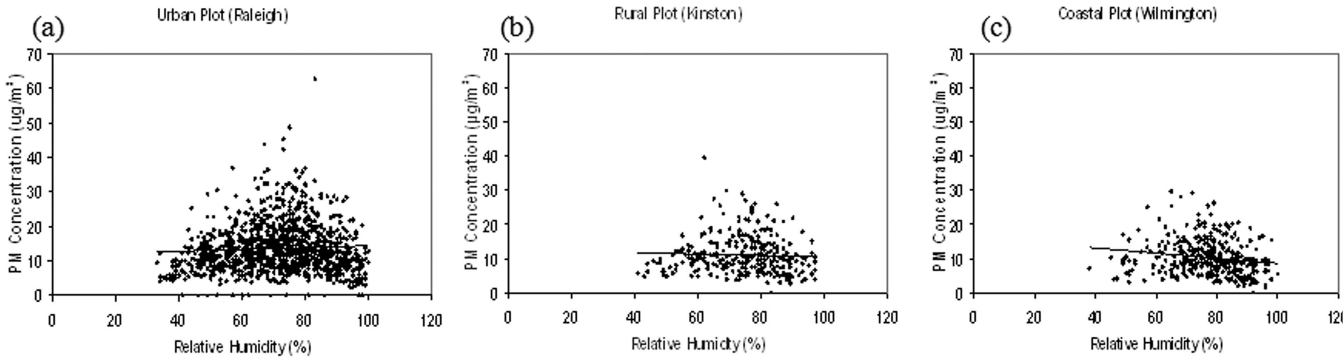

Figure 3. RH vs. $PM_{2.5}$ concentration at (a) Raleigh (urban), (b) Kinston (rural), and (c) Wilmington (coastal).

the hog facilities on $PM_{2.5}$ concentrations can be seen at some sites. For example, higher $PM_{2.5}$ average concentrations were found from a southeasterly flow at Raleigh (urban), which corresponds to Raleigh's orientation to the hog facilities. High $PM_{2.5}$ concentrations at Kinston (rural) were from the southwest and west directions, which corresponded exactly to Kinston's orientation to most of the hog facilities. The highest average concentrations at Fayetteville were found from the southeast direction, rather than the east from where the emissions of hog facilities come. The weak correlation between the $PM_{2.5}$ concentrations and the east wind direction at Fayetteville was likely because fewer measurements were available at this site and the easterlies were not the prevailing winds during those days with observations, which would make this site more susceptible to other local sources and variability. At the other two rural sites (i.e., Goldsboro and Kenansville), relatively homogeneous correlation between $PM_{2.5}$ concentrations and cardinal directions was found. High $PM_{2.5}$ average concentrations at Goldsboro were from the southeast, southwest, west, and north directions, with the peak concentrations coming from the southeast. The $PM_{2.5}$ concentrations range from 2.7 to 31.4 $\mu g \cdot m^{-3}$, with an average of 10.8 $\mu g \cdot m^{-3}$ at Kenansville, which is very high for a small rural town. This indicates the impact of the hog facilities. The two coastal sites (i.e., Jacksonville and Wilmington) had higher concentrations from the southwest and west directions, indicating the impact of emissions from the state of South Carolina. High correlation was also found for the east direction at Jacksonville and the northwest direction at Wilmington.

$PM_{2.5}$ MODELING RESULTS

Figure 7 shows the observed and predicted average total inorganic $PM_{2.5}$ concentrations and its composition at three sites: Fayetteville, Kinston, and Raleigh ("total inorganic $PM_{2.5}$ or total inorganic PM" is defined as the sum of the four major inorganic constituents: ammonium, chloride, nitrate, and sulfate). The predicted values were obtained under the conditions with median initial total PM species concentration, median RH, and median T, as shown in Table 2. The observations at all three sites showed that sulfate has the largest contribution (approximately two-thirds of the total observed inorganic aerosol), followed respectively by ammonium, nitrate, and chloride. The simulation results from ISORROPIA generally agree well with observed $PM_{2.5}$ in terms of both magnitude and composition. Compared with observed total inorganic $PM_{2.5}$ concentration, ISORROPIA underestimated by 0.50–0.75 $\mu g \cdot m^{-3}$ (8.7–12.5%) at Fayetteville and Kinston, and overestimated the observed values by 0.37 $\mu g \cdot m^{-3}$ (6.9%) at Raleigh. At all three sites, sulfate had the largest contribution, followed respectively by ammonium, nitrate, and chloride. The ammonium concentration at Kinston and Fayetteville was underpredicted by approximately 0.1 $\mu g \cdot m^{-3}$ (~7.4%) and that at Raleigh was overpredicted by the same value (7.7%). The largest differences between observed and predicted values were in the nitrate concentration. It was underpredicted by 0.39 $\mu g \cdot m^{-3}$ at Fayetteville and 0.54 $\mu g \cdot m^{-3}$ at Kinston (48 and 59%, respectively). The nitrate concentration predicted at Raleigh was 0.25 $\mu g \cdot m^{-3}$ (37%) greater than the observed nitrate concentrations. The observed chlorine concentrations were nearly zero whereas

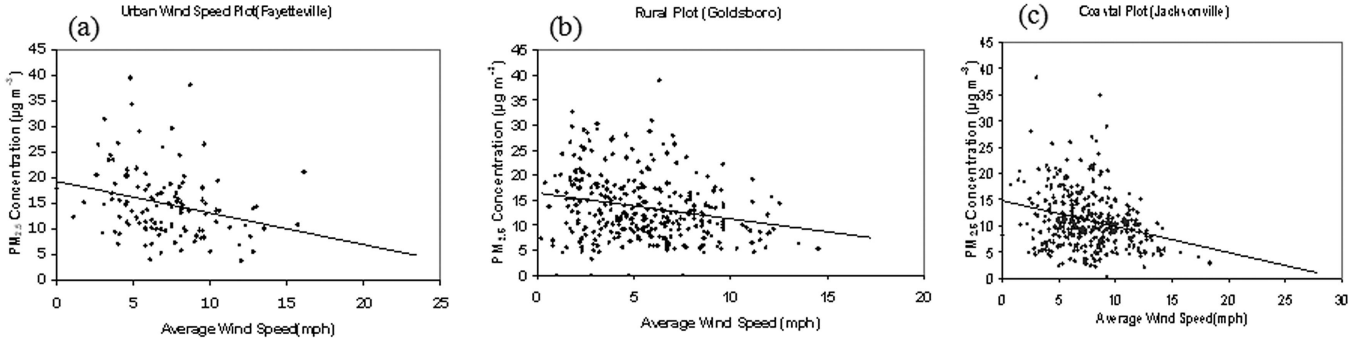

Figure 4. Wind speed vs. $PM_{2.5}$ concentration for (a) Fayetteville (urban), (b) Goldsboro (rural), and (c) Jacksonville (coastal).

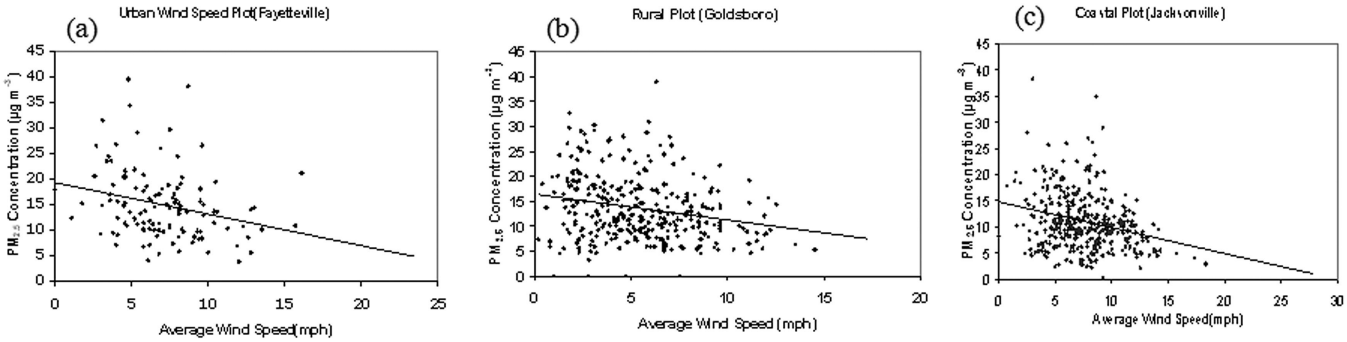

Figure 5. T vs. $PM_{2.5}$ concentration for (a) Raleigh (urban), (b) Kenansville (rural), and (c) Wilmington (coastal).

the predicted chlorine concentrations at the three sites were less than 0.1 $\mu g \cdot m^{-3}$. At each site, the predicted pH and aerosol water concentrations were 7.53–7.56 and 5 $\mu g \cdot m^{-3}$, respectively. The model gave the best agreement against observations at Raleigh among the three sites.

Figure 8 shows the predicted total inorganic $PM_{2.5}$ concentration at the maximum initial pollutant concentrations at each site under the three different meteorological settings. The maximum observed values are also plotted for comparison. For the median RH/median T and the maximum RH/minimum T conditions, the predicted total $PM_{2.5}$ inorganic aerosol concentrations ranged from 26 to 50 $\mu g \cdot m^{-3}$ at the three sites, which consistently overpredict the observed maximum concentrations (15–18 $\mu g \cdot m^{-3}$) at all sites. The predicted total inorganic $PM_{2.5}$ concentration increased as the urban development of the area increased (Kinston [rural]), Fayetteville [small city], Raleigh [large city]). These differences are because of differences in the predicted particulate nitrate concentration, which was a factor of 2 and 3 higher at Fayetteville and Raleigh, respectively, than that at Kinston. The predicted particulate ammonium concentration was higher by 32 and 80% at Fayetteville and Raleigh, respectively, because of formation of NH_4NO_3. With the higher sulfate concentrations at Kinston and Raleigh, the aerosol was much more acidic at these sites (with pH values of 4.5–4.8), whereas that at Fayetteville was more neutral (6.8). The predicted total inorganic aerosol concentrations ranged from 17.23 to 19.09 $\mu g \cdot m^{-3}$ at the three sites under the minimum RH/maximum T condition. Such a condition favors evaporation of nitrate and water, resulting in zero nitrate and water concentration in the aerosol phase. The aerosol consisted of primarily $(NH_4)_2SO_4$. The differences in predicted total inorganic aerosol concentrations among these sites were thus much smaller.

Table 3. The slope, *y*-intercept, and linear fit R^2 value from the total $PM_{2.5}$ vs. ammonium $PM_{2.5}$ plots for Kinston, Fayetteville, and Raleigh.

Site Name	Slope	*y*-Intercept	R^2
Fayetteville	0.0841	0.1758	0.591
Kinston	0.0168	1.21	0.011
Raleigh	0.0995	−0.05	0.712

A similar plot is shown at the minimum pollutant concentrations at each site in Figure 9. The model underestimated the observed minimum concentrations by less than 1 $\mu g \cdot m^{-3}$ at each site. Under the median RH/median T and the minimum RH/maximum T conditions, the total inorganic $PM_{2.5}$ concentrations were the same and they consisted of sulfate salts only. The nitrate concentrations were either zero or negligible. Under the maximum RH/minimum T conditions, some nitrate formed. The total PM species concentrations predicted at the three sites ranged from 1.01 to 1.11 $\mu g \cdot m^{-3}$. Under this condition, the urban areas were characterized by 20% more sulfate than the rural site, but the rural site (i.e., Kinston) had slightly more nitrate, ammonium, and chloride, resulting in total $PM_{2.5}$ concentration that is slightly higher than that at Fayetteville but lower than that at Raleigh.

CONCLUSIONS

The concentrations and trends of $PM_{2.5}$ in eastern North Carolina were studied with data analysis and an aerosol thermodynamic box model that predicted the gas/particle partitioning of PM. The unique emission fluxes of pollutants (e.g., NH_3) from the hog industry and their impacts on PM concentrations make this region a unique environment to understand the role of these emissions in PM formation.[20] The major constituents of $PM_{2.5}$ from the greatest to the least are OC, sulfate, ammonium, nitrate, and EC. Higher $PM_{2.5}$ concentrations tended to occur between 60 and 90% RH, with this effect being more pronounced in urban areas. There was a positive relationship between T and $PM_{2.5}$ concentrations, and a negative relationship between wind speed and $PM_{2.5}$ concentrations. The box-whisker plots of wind direction demonstrate that there is a connection between hog facility density and $PM_{2.5}$ concentration, but with the limited data, these concentrations could not be attributed to any specific pollutant.

ISORROPIA was used to simulate the gas/particle partitioning and the total inorganic aerosol concentration at three sites in eastern North Carolina. The model predictions showed that the major predicted constituents of inorganic aerosols were sulfate, ammonium, and nitrate, which agreed with the overall measurements. The predicted average total inorganic concentrations were slightly (<1 $\mu g \cdot m^{-3}$) lower than the observations. Although the model predicts the concentrations of sulfate

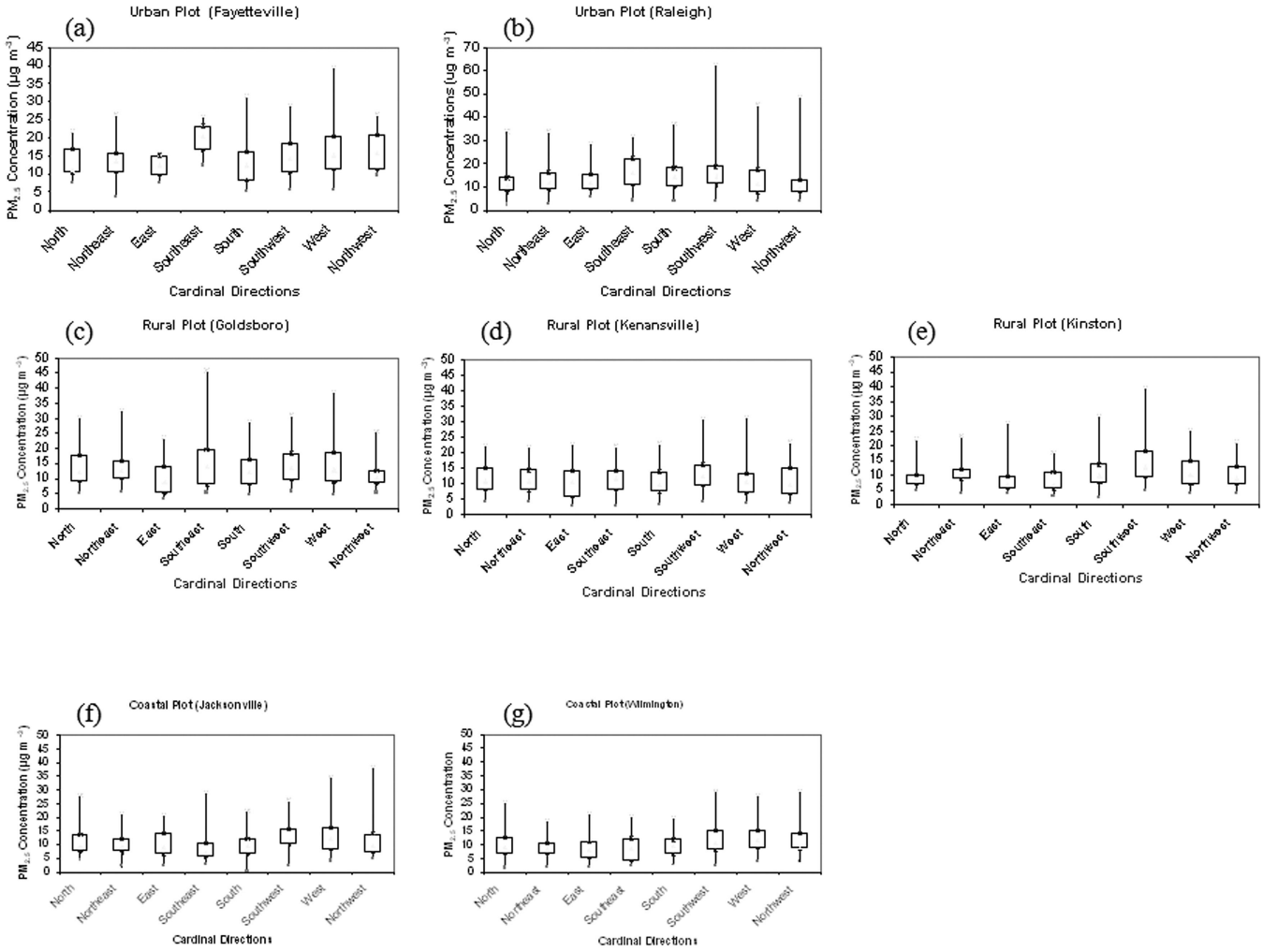

Figure 6. Wind direction box-whisker plots for (a) Fayetteville (urban), (b) Raleigh (urban), (c) Goldsboro (rural), (d) Kenansville (rural), (e) Kinston (rural), (f) Jacksonville (coastal), and (g) Wilmington (coastal).

and ammonium that are in good agreement with observations, it tends to underpredict the observed particulate nitrate concentrations by 0.22 $\mu g \cdot m^{-3}$ (27.5%) at all three sites. The simulation results were sensitive to initial total PM concentrations and meteorological conditions, with the highest secondary PM formation occurring under the condition with maximum initial total PM concentrations, maximum RH, and minimum T.

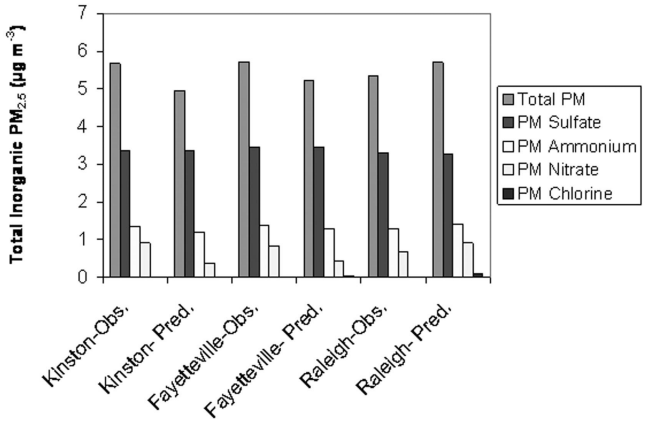

Figure 7. Observed and predicted total inorganic $PM_{2.5}$ concentrations at Kinston, Fayetteville, and Raleigh, NC, under median RH and T conditions.

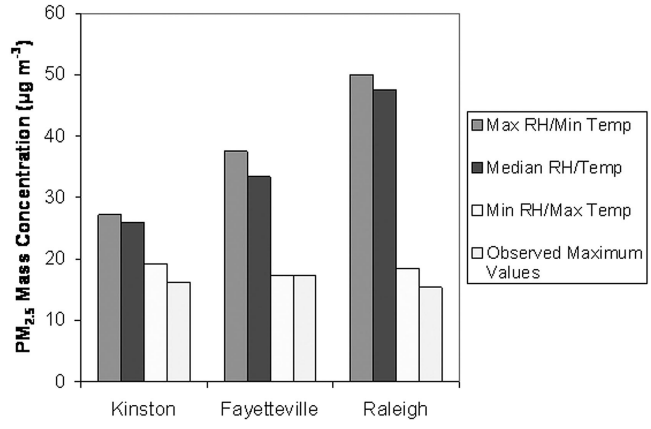

Figure 8. Predicted total inorganic $PM_{2.5}$ concentrations under three meteorological conditions and maximum observed total inorganic $PM_{2.5}$ concentrations at Kinston, Fayetteville, and Raleigh, NC.

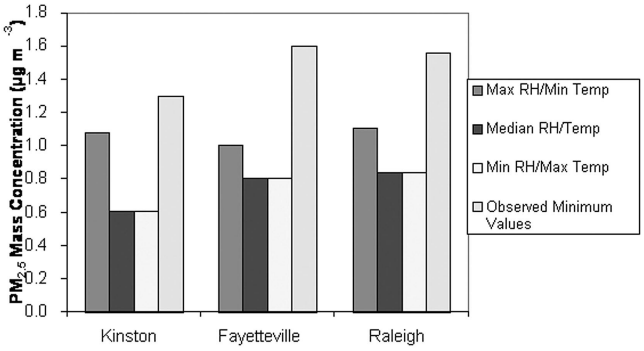

Figure 9. Predicted total inorganic $PM_{2.5}$ concentrations under three meteorological conditions and minimum observed total inorganic $PM_{2.5}$ concentrations at Kinston, Fayetteville, and Raleigh, NC.

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