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07/15/96

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Main project #: PE #:

Project unit: E & A SCI Unit code: 02.010.140
Project director(s):
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Sponsor/division names: GA DEPT NATURAL RESOURCES /
Sponsor/division codes: 300 / 031

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PROJECT ADMINISTRATION DATA

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AMENDMENT 3 EXTENDS PERIOD OF PERFORMANCE THROUGH DECEMBER 31, 1996.

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OFFICE OF CONTRACT ADMINISTRATION

NOTICE OF PROJECT CLOSEOUT

Closeout Notice Date 01/17/97

Project No. G-35-803

Center No. 10/31-6-T5429-0A0

Project Director RODGERS M O

School/Lab E & A SCI

Sponsor GA DEPT NATURAL RESOURCES/

Contract/Grant No. 771-390279 Contract Entity GTRC

Prime Contract No.

Title COOPERATIVE RESEARCH FOR AIR QUALITY IN GEORGIA

Effective Completion Date 961231 (Performance) 961231 (Reports)

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Submitted

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Final Report of Inventions and/or Subcontracts	Y	
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Comments

Subproject Under Main Project No.

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Emissions and Urban Airshed Modeling Issues for Atlanta, Georgia

Final Report (GTRC contract #771-390279)

Submitted to
Air Protection Branch
Environmental Protection Division
Georgia Department of Natural Resources
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I. Executive Summary

In January 1994, we proposed several projects related to emissions and air quality modeling issues for Atlanta, Georgia. In the Spring of 1994, we received a contract (GTRC contract # 771-390279) from the Georgia Department of Natural Resources, Environmental Protection Division, Air Protection Branch (GA DNR) to complete seven tasks. The seven tasks were:

- 1) Emission Inventory Preparation
- 2) Urban Airshed Model (UAM) Comparison with Observations
- 3) Inverse Method Development
- 4) Inverse Method Validation
- 5) Preliminary Control Strategy
- 6) Inverse Runs using Observations from Southern Oxidants Study (SOS) 1992 Atlanta Intensive
- 7) Activity Factor / Emission Factor Improvement

By mutual agreement (section 4, Schedule of the original proposal), these seven tasks were to be completed according to the following schedule (the number in parentheses corresponds to the task number above):

April-June, 1994

- ◆ prepare 1992, 1990 (assist GA DNR), and SIP (assist GA DNR) emission inventories (1);
- ◆ use inverse method to determine preliminary control strategy for 1999 (with caveats) (5);
- ◆ conduct "pseudo-data" inverse studies with UAM to develop method (3).

July-September, 1994

- ◆ complete "pseudo-data" studies (4);
- ◆ run UAM for 1992 episode, compare to observations (2);
- ◆ use test tracer to validate inverse method (4).

October-December, 1994

- ◆ run inverse with UAM using SOS observations, compare to 1992 emissions inventory (6).

January-March, 1995

- ◆ improve activity factors and emission factors, target uncertainties in emissions inventory (7);
- ◆ use inverse method to revise preliminary control strategy for 1999 (5).

Key findings from our research are summarized below and discussed in detail in section II of this report. Further, although we cannot provide a preliminary control strategy at this time, we have initiated an expanded research program to address this topic. The expanded program grew out of our findings related to the control strategy task. Finally, in the course of our research, we conducted several relevant experiments not specifically listed in the original proposal. These are also discussed here under an eighth task heading of "Other Research."

1. Emission Inventory Preparation

The Emissions Preprocessing System - version 2 (EPS2) is a set of computer programs to process emissions for use in the Urban Airshed Model (UAM). In collaboration with the program developers at EPA, we ported, debugged, and updated this code to our UNIX machine. We then shared our working system with the GA DNR.

For specific applications, we provided the GA DNR (directly or through their agent, Systems Applications International) with a variety of emission inventory products including digitized road links for the Atlanta UAM domain, mobile and point source emissions for August 9-10, 1992, and satellite-derived land-use data for the Atlanta UAM domain.

2. UAM Comparison with Observations

We conducted a series of intercomparisons between ambient measurements, emission estimates, and UAM simulations to estimate the quality of the emission estimates and the uncertainty in UAM simulations for Atlanta. Our key findings were:

- ◆ The ambient ratios of AHC/NO_x and CO/NO_x are higher than the corresponding emission ratios by 20% and 73% respectively.
- ◆ The ambient concentrations of AHC, NO_x, and CO are greater than the corresponding UAM concentrations by 14%, 40% and 58%, respectively.
- ◆ UAM calculated concentrations of isoprene are 3.9 to 8.6 times less than the observed ambient isoprene concentrations.

This last finding, when reported to the developers of the Biogenic Emissions Inventory System (BEIS), was an impetus for them to develop a second generation BEIS (BEIS2).

3. Inverse Method Development

Using ambient observations, an inverse method was used to estimate the emissions needed to minimize the differences between UAM predicted and ambient observed concentrations. We implemented a specific form of the inverse method called the Kalman Filter. The filter was adapted to work with the regulatory version of the UAM to ensure compatibility between the research and regulatory communities. The code is a mixture of FORTRAN-77 and UNIX Korn shell. It was written for a Hewlett Packard 9000 series 735 machine; thus, it is fully compatible with GA DNR's UNIX platform.

4. Inverse Method Validation

The application of the Kalman Filter to the UAM was validated using pseudo-data. Pseudo-data are time series of model generated chemical concentrations from a model grid cell at the same location as a real observation station. By intentionally introducing an error into the emissions, we demonstrated that the inverse method worked by using the difference between the model generated concentrations and the pseudo-data to iterate back to the correct emissions. Due to the success of this demonstration and the lack of a suitable tracer in Atlanta's real atmosphere, we concluded that this test was sufficient.

5. Preliminary Control Strategy

We found that our proposed method for determining a control strategy could not work for the question of ozone pollution. It required ozone to be linearly related to its precursors. We found that we needed a method that could deal with the nonlinear relationship between ozone and its precursors, and address the range of time scales critical to photochemical transport on the urban scale. We have since identified a method used extensively in engineering optimization problems. We are collaborating with a group in the School of Industrial and Systems Engineering to further develop this program.

6. Inverse Runs using Observations from SOS 1992 Atlanta Intensive

As a highly reactive precursor, the role of isoprene is critical to understanding ozone pollution. Observed concentrations of isoprene from the SOS 1992 Atlanta Intensive, the UAM, and the inverse method were used to estimate biogenic isoprene emissions and improve the simulation of isoprene concentrations. Five simulations were compared:

- ◆ Base case UAM using BEIS (current regulatory model).
- ◆ Base case UAM using BEIS2 (proposed regulatory model).
- ◆ Inverse corrected run using BEIS.
- ◆ Inverse corrected run using BEIS2.
- ◆ Inverse corrected run using BEIS2 and allowing rural and urban biogenic isoprene emissions to change independently.

These are the results showing the bias and gross error of predicted isoprene concentrations relative to observations, and the average change in isoprene emissions relative to BEIS.

<i>Biogenic Emissions</i>	<i>Bias</i>	<i>Gross Error</i>	<i>Average Emissions Change Relative to BEIS</i>
BEIS	-85%	85%	---
BEIS2	-34%	61%	~ 4X
Inverse Corrected BEIS	-8%	60%	~ 5X
Inverse Corrected BEIS2	-9%	57%	~ 6X
Inverse Corrected BEIS2 (Independent Urban & Rural)	-5%	42%	~ 11X

These results show that BEIS underpredicts isoprene emissions across the whole UAM domain by at least a factor of 4 and as much as a factor of 11. Further, while BEIS2 is an improvement over BEIS, the last case indicates that the spatial distribution of biogenic emissions in BEIS2 is insufficient. We present a biogenic isoprene emission inventory which, when used with the UAM, reduces the error between isoprene observations and predictions.

7. Activity Factor / Emission Factor Improvement

After showing that the regulatory modeling system of BEIS and UAM grossly underpredicts isoprene concentrations, we collaborated with EPA scientists to produce the UAM version of the Biogenic Emissions Inventory System - version 2 (BEIS2). We conducted several numerical computations using the new code to investigate its impact on the prediction of isoprene and ozone concentrations, and its effect on the design of ozone control strategies. Using BEIS2, we found:

- ◆ The biogenic emission contribution to the total VOC within the Atlanta UAM domain changed from 55% (29% isoprene and 26% monoterpenes) using BEIS, to 76% (59% isoprene and 17% monoterpenes) using BEIS2.
- ◆ The simulated maximum ozone concentration changed from 129 ppbv with BEIS, to 169 ppbv with BEIS2. The total ozone production increased by 14% with the new biogenic emissions.
- ◆ The Atlanta region changed from being VOC-limited under BEIS, to NO_x-limited under BEIS2.

Using survey data collected during the SOS 1992 Atlanta Intensive, we calculated emissions factors for two area sources: automobile refinishing and graphic arts.

- ◆ For automobile refinishing, we found a factor of two between the EPA's per employee factor (3,519 lbs/employee/year) and the factor derived from the surveys (1,656 lbs/employee/year).
- ◆ For graphic arts, the emissions from Fulton County appear to be different than those from the other counties. Our emission factor agrees with the EPA's per capita factor only when we exclude Fulton County.
- ◆ For both automobile refinishing and graphic arts, the 5 days/week operating schedule used in the current emission inventory is a valid assumption.

8. Other Research

About 90% of point source NO_x emissions are from four power plants: Bowen, McDonough, Wansley, and Yates. With an experiment to estimate the sensitivity of ozone to these sources, we found:

- ◆ Removing emissions from McDonough causes an overall increase in O₃.
- ◆ Removing emissions from Bowen reduces O₃.

Comparing runs using meteorology from a diagnostic meteorology model with standard meteorological data, a diagnostic meteorology model with enhanced meteorological data (lidar generated mixing heights), and an advanced prognostic meteorology model (from the Systems Applications International Mesoscale Model), for the August 10, 1992 simulation using identical emissions, we found:

- ◆ The maximum O₃ concentration predicted using the prognostic model (201 ppbv) was 19% higher than the maximum O₃ predicted with the standard diagnostic model (169 ppbv). The total ozone production increased 14%.
- ◆ The maximum O₃ predicted by the enhanced diagnostic model (131 ppbv) was 22% lower than the maximum O₃ predicted by the standard diagnostic model (169 ppbv). The total ozone production decreased by 3%.

These results illustrate the considerable sensitivity of the ozone simulation to uncertainties in the meteorology.

II. Emissions and Urban Airshed Modeling Issues for Atlanta, Georgia

In our original proposal, we agreed to complete work according to the following schedule (the number in parentheses following each task indicates the subsection below in which the task is addressed):

April-June, 1994

- ◆ prepare 1992, 1990 (assist GA DNR), and SIP (assist GA DNR) emission inventories (1);
- ◆ use inverse method to determine preliminary control strategy for 1999 (with caveats) (5);
- ◆ conduct "pseudo-data" inverse studies with UAM to develop method (3).

July-September, 1994

- ◆ complete "pseudo-data" studies (4);
- ◆ run UAM for 1992 episode, compare to observations (2);
- ◆ use test tracer to validate inverse method (4).

October-December, 1994

- ◆ run inverse with UAM using SOS observations, compare to 1992 emissions inventory (6).

January-March, 1995

- ◆ improve activity factors and emission factors, target uncertainties in emissions inventory (7);
- ◆ use inverse method to revise preliminary control strategy for 1999 (5).

1. Emission Inventory Preparation

a. Emissions Preprocessing System (EPS2)

Gridded emissions of hourly resolved, chemically speciated hydrocarbons, NO, NO₂ and CO were prepared using the Emission Preprocessor System - Version 2 (EPS2). EPS2 was used to be consistent with the EPA guidelines followed by the Georgia Department of Natural Resources (GA DNR) in developing their State Implementation Plan for Ozone Non-attainment in Atlanta, Georgia (SIP).

A considerable amount of time and effort was spent on getting EPS2 up and running. Working in close contact with the Environmental Protection Agency (EPA) personnel that were responsible for overseeing the development and the application of EPS2, we identified errors within the original code and developed solutions for those bugs. The updated and improved EPS2 code that our group produced was shared with the GA DNR.

b. Emission Inventory Products

In addition to the improved EPS2 code, GA DNR (or their agent, Systems Applications International) received from our group the following emission inventory products:

- ◆ Digitized road links covering all interstate and principal roads within the Atlanta UAM domain.
- ◆ Daily mobile emissions of VOC, NO_x and CO by vehicle type, for all road links, for a 1990 typical summer day, and for August 9 and 10, 1992.
- ◆ Daily mobile emissions of VOC, NO_x and CO by vehicle type, for all road links, for July 29, 30, 31, and August 1 of 1987 and July 7 and 8 of 1988. (GA DNR requested these to help quality assure a duplicate effort by SAI.)
- ◆ Day-specific point source emissions for August 9 and 10 of 1992.
- ◆ Satellite-derived land-use data for the Atlanta UAM domain.

2. UAM Comparison with Observations

Once the emission inventories were created, the next task was to estimate how well the emission estimates represented the atmospheric chemical conditions in the Atlanta area. To accomplish this, we used two different approaches. In one method, we compared ambient measurements obtained during the Atlanta field studies of 1990 and 1992 with the corresponding emission inventory estimates. In the second method, we compared ambient measurements with simulated concentrations obtained using the Urban Airshed Model (UAM).

These findings were presented in more detail at the *Air and Waste Management Association International Conference on the Emission Inventory: Applications and Improvements* in November, 1994. A copy of the presented paper is included in Appendix A.

a. Anthropogenics

For anthropogenic emissions, we compared observed ratios of anthropogenic hydrocarbons (AHC) to oxides of nitrogen (NO_x) and observed ratios of carbon monoxide (CO) to NO_x to corresponding ratios derived from emission estimates. The results show that:

- ◆ The ambient ratios of AHC/NO_x and CO/NO_x are higher than the emission ratios by 20% and 73% respectively

Comparing UAM predicted concentrations of AHC, NO_x, and CO to observed concentrations, we found that:

- ◆ The ambient concentrations of AHC, NO_x, and CO are greater than the corresponding UAM concentrations by 14%, 40% and 58%, respectively;

The variability shown by the different comparisons suggests that a possible source of the discrepancies is the spatial distribution of anthropogenic emissions.

b. Biogenics

For biogenic emissions, we focused on isoprene, the most reactive hydrocarbon in the Atlanta chemical mix. We compared observed isoprene concentrations at several sites with UAM simulated concentrations. The comparison showed that the observed isoprene concentrations were 3.9 to 8.6 times higher than the UAM concentrations. We also produced a UAM simulation in which the isoprene emissions were increased by a factor of 5 throughout the day over the whole domain. This isoprene-enhanced simulation showed more realistic isoprene concentrations during the morning and early afternoon hours, but unrealistic isoprene levels by late evening. Another difficulty with the isoprene-enhanced simulation was the increase in ozone concentrations. A comparison using 15 ozone monitoring stations showed that the isoprene-enhanced case generates about 40% more ozone than the observed one. The results can be summarized as follows:

- ◆ The isoprene emissions are substantially underrepresented in the inventories.
- ◆ The magnitude of the isoprene emission underestimation depends on the time of day.

3. Inverse Method Development

The inverse application used in this research is an amalgamation of FORTRAN and UNIX Korn shell programs. The method was developed on an Hewlett Packard 9000, Series 735 workstation running the HP-UX UNIX operating system. The latest version (5/24/95, included as Appendix B) is a result of continuous program improvement. To facilitate the use of the inverse method for regulatory guidance, this application uses the latest regulatory version of the Urban Airshed Model - IV (US EPA, 1992).

a. Background

As discussed in subsection 2, the UAM adequately predicts ozone despite a gross underprediction in ozone precursors, particularly isoprene. Thus, although the model reproduces the ozone observations, it may not be getting that correct ozone prediction for the right reasons. Without an accurate simulation of the mechanisms that create and remove ozone in the atmosphere, it may be virtually impossible to create effective control strategies. However, if predicted ozone precursor concentrations match observed ozone precursor concentrations and the model is formulated correctly, then the predicted ozone should match the observed ozone for the right reasons. In other words assuming the UAM is a valid model,

- ♦ predicting the correct concentrations of ozone is a necessary but not sufficient criteria for validating a simulation; or
- ♦ predicting the correct concentration of ozone precursors is a necessary and sufficient criteria for validating a simulation.

Current air quality models mathematically simulate the relationship between one or more dependent variables (e.g. ozone) and one or more independent variables (e.g. emissions and meteorology). The model may be thought of as a *transformation function* whereby the model input is transformed into the model output. For example, consider this simple model for calculating ozone time series at a monitoring location (Zurbenko et al, 1994):

$$O_t = W_t + [aT_{t+\Delta t} + b] + \epsilon_t \quad (1)$$

where

O_t is the ozone concentration at time t

W_t represents the short term variation in O_t (the noise)

ϵ_t is the long-term trend component due to changes in ozone precursor emissions

$[aT_{t+\Delta t} + b]$ is the temperature dependent component (a and b are constants)

Equation (1) may also be represented in a more compact matrix notation as

$$\mathbf{d} = \mathbf{Gm} \quad (2)$$

where

\mathbf{d} is a vector of data, O_t

\mathbf{m} is a vector of model parameters, W_t , $[aT_{t+\Delta t} + b]$, and ϵ_t

\mathbf{G} is an 1 by 3 *transformation matrix* which in this case is trivially, $[1 \ 1 \ 1]$

This latter format is useful when there are many data points, many model parameters, or a complex transformation matrix. This is the format we will employ in this work.

With this model, the independent variables (W_t , $[aT_{t+\Delta t} + b]$, ϵ_t) are mathematically transformed into the dependent variable (O_t). As long as the values of W_t , $[aT_{t+\Delta t} + b]$, and ϵ_t are known, a unique solution, O_t , can be found. This predicted value can then be compared to an observed value. However, if two or more of the dependent variables

are unknown or uncertain, there is an infinite combination of W_t , $[aT_{t+\Delta t} + b]$, and ϵ_t such that any observed value may be predicted by the model. Thus, even with the wrong input variables, it is possible to calculate the correct solution! Models with more independent variables (there are literally millions of input parameters for the Urban Airshed Model) inevitably compound this error.

We will use an *inverse method* to reduce the probability of the Type II error described above from occurring. Inverse methods are typically used when a valid model exists to relate independent variables to dependent variables, but the independent variables are not well known. To work, the inverse method requires an independent set of observations of the values the model is trying to predict. In the example above, even if W_t , $[aT_{t+\Delta t} + b]$, and ϵ_t are all unknown, using an inverse technique with a series of observations, O_t^{obs} , it may be possible to determine the correct values of W_t , $[aT_{t+\Delta t} + b]$, and ϵ_t needed to reproduce the observation set. Since there are three unknowns, W_t , $[aT_{t+\Delta t} + b]$, and ϵ_t , at least three observations are needed to solve for all three variables. A unique solution can be found with exactly three independent observations. If there are more than three observations, a best fit - least squares solution may be found.

Inverse methods have been successfully applied to many fields including medical imaging, missile guidance, and underground tomography. In recent applications in the atmospheric sciences, inverse methods were used to estimate the continental-scale emissions of CFCs (Hartley and Prinn, 1993), area-source emissions from downwind concentration measurements (Lehning et al, 1994), and the temporal and spatial distribution of CO emissions in the Los Angeles basin (Mulholland and Seinfeld, 1995). This study is the first to apply the inverse method to estimate the emissions of the relatively short-lived species important to urban oxidants formation.

The Urban Airshed Model is much more complicated than the simple model shown above. Nevertheless, the idea and technique behind the inverse method may be similarly applied. We used a particular form of the inverse method called the *Kalman filter*.

b. The Kalman Filter

By adjusting the emissions, the Kalman filter form of the general inverse method may be used to reconcile emission based, model predicted concentrations of pollutants located in time and space (χ^{model}) with corresponding observations (χ^{obs}). As implemented here, the differences between observations and model-generated concentrations ($\Delta\chi = \chi^{obs} - \chi^{model}$) are used to calculate the requisite change in emissions (ΔE) from different sources,

$$\Delta E = G\Delta\chi \quad (3)$$

where

$$G = CP^T(PCP^T + N)^{-1} = \text{"Gain matrix"} \quad (3a)$$

and

$$P = [P_{ij}] = \frac{\partial \chi_i}{\partial E_j} = \text{"Jacobian matrix"} - \text{Effect of emissions source } j \text{ on observation } i \quad (3b)$$

$$N = [N_i] = [\sigma_i^{obs^2}] = \text{"Noise matrix"} - \text{Covariance matrix (diagonal) of errors in observation } i \quad (3c)$$

$$C = [C_j] = [\sigma_j^{E^2}] = \text{"Covariance matrix"} - \text{Covariance matrix (diagonal) of errors in emission } j \quad (3d)$$

Notice how equation (3) is similar in form to equation (2). Through a transformation matrix, here called the Gain matrix, differences between observations and model predictions are transformed into differences between the emissions needed to reproduce the observations and the emissions needed to produce the model predictions.

The covariance matrix of emission errors may be updated after each iteration,

$$\mathbf{C}_{\text{new}} = \mathbf{C}_{\text{old}} - \mathbf{GPC}_{\text{old}} \quad (4)$$

c. Time Dependence Development

Previous work (Hartley, 1992; Hartley and Prinn, 1993) has demonstrated the use of the Kalman Filter to deduce the emissions for CFC-11. However, there is an important difference that must be addressed when inverting for the shorter lived gases relevant to urban ozone. This difference is that the urban pollution gases (e.g. NO_x, VOCs) all have diurnal cycles associated with their different sources; whereas, CFC-11 has fairly constant emissions. To that end, our first task was to develop the Kalman filter for use as an inverse method to determine time varying emissions.

In order to be able to clearly identify the components of the inverse problem and to affordably run a wide variety of tests, we chose to use a simplified analytical regional scale model based on the Effectively Sampled Region (ESR) model (Prinn, 1988). In this simplified model we do not use real observations, but rather model generated observations or pseudo-data. The goal of this initial work was to develop a method which could be applied to the more complex atmospheric models, like the UAM, to determine the time-varying sources and sinks of the photochemically active gases.

The reason we needed to address the time varying issue explicitly is that the standard recursive Kalman filter converges on a solution by continuously narrowing the size of the emission adjustments. In order to allow for the necessary adjustments when the emissions are changing, we used an adaptive filter. Adaptive filters have been widely used in the engineering fields to handle changing state variables (Sastri, 1995). After running tests with our simplified model using the adaptive filter, we found this modification greatly improved the inversion for time varying sources.

Even with this adaptive filter, an additional problem remained. A time lag was introduced as a result of the detection criterion that allows the filter to adapt. To eliminate, or at least minimize, this time lag, several methods were considered. We concluded that the most accurate method was to iterate at each time step until the model was in reasonable agreement with observed concentrations. The iterative process eliminated the propagation of a poor initial guess of the emissions and allowed the inverse method to determine the correct time varying sources.

This study led to a paper submitted to the *Journal of Geophysical Research*. A copy is included in appendix A.

d. Application of the Kalman Filter to the UAM

The remainder of this section describes how we have calculated or obtained each of the matrices for the Kalman filter using the Urban Airshed Model (UAM) for Atlanta, Georgia. The general application is shown schematically in Figure 3.1.

The initial step involves calculating the Jacobian matrix, \mathbf{P} . Each element in this matrix is the sensitivity of a pollutant concentration at a particular place and time to an emission of specific type and magnitude. We used the UAM to directly calculate these factors by assuming,

$$\frac{\partial \chi_i}{\partial E_j} = \frac{\Delta \chi_i}{\Delta E_j} = \frac{\chi_i(E) + \chi_i(E + \delta E_j) - \chi_i(E)}{\delta E_j} \quad (5)$$

where

$\chi_i(E)$ = UAM predicted air pollutant concentration at station, i , given emissions E

δE_j = a small perturbation in some emission source, j (we used $\delta E_j = 10\% E_j$)

Figure 3.2 (a) shows the sensitivity of NO_x concentrations at Georgia Tech to a 10% change in NO_x emissions over the whole UAM domain. Figure 3.2 (b) is a similar plot showing the sensitivity of anthropogenic

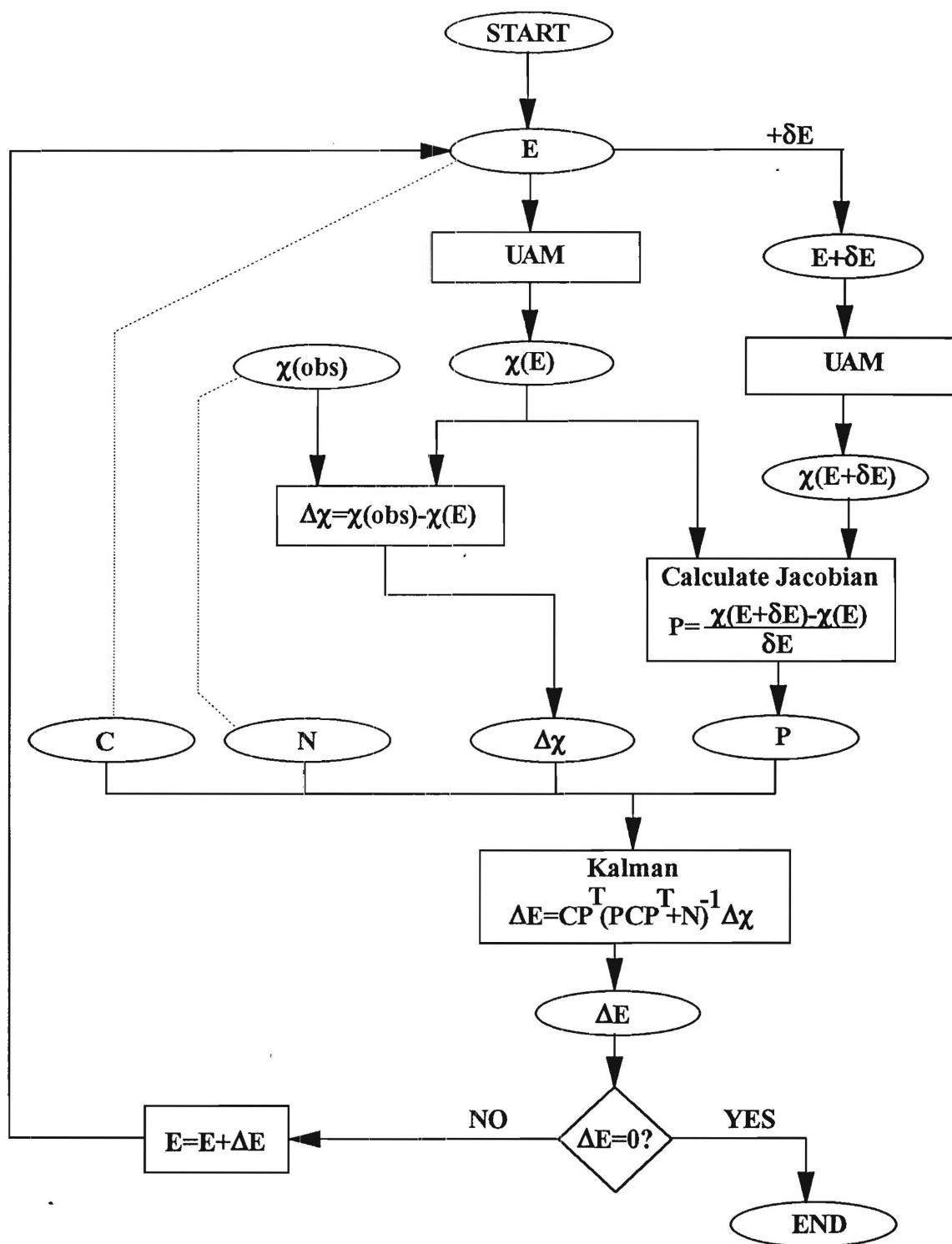


Figure 3.1. Schematic diagram of the application of the Kalman filter to the UAM.

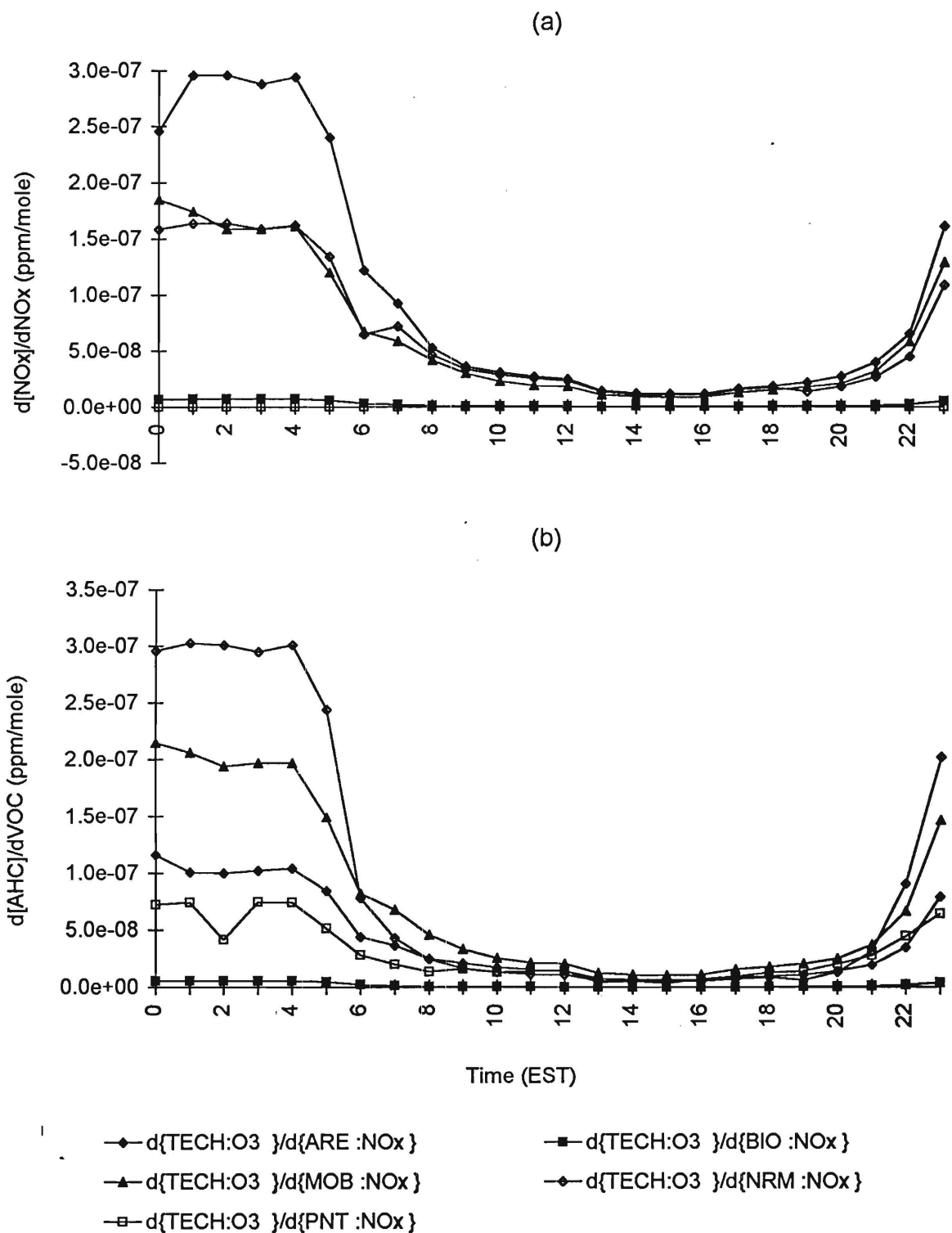


Figure 3.2. Ozone precursor sensitivity to (a) NO_x and (b) VOC emissions.

hydrocarbon (AHC) concentrations to VOC emissions. On a 1 hour time scale, the primary Jacobian elements (e.g. the change in NO_x concentrations due to a change in NO_x emissions) are insensitive to variations in the concentration of other species, the initial concentration of the species of interest, and the size of the emissions perturbation, δE_j . Thus, at the Georgia Tech site, the profiles shown in figure 3.2 will remain constant regardless of any changes in emissions. The secondary Jacobian elements (e.g. the change in O₃ concentrations due to a change in VOC emissions) are sensitive to variations in all of these parameters and thus, must be recalculated each time one of them changes. This latter characteristic stalled our efforts to develop a control strategy using the inverse method. It is discussed in more detail in subsection 5.

N , the noise matrix, is composed of the squares of the errors in observation measurements, σ^{obs} . Certainly this includes instrument error, but it may also include the more difficult to quantify, uncertainty of a particular site's ability to represent the air quality of a larger region (or grid cell) over space and time. If it is believed that a particular observation is not representative of the local air quality, its relative importance in the Kalman filter calculation may be diminished by increasing the noise component. When using synthetic observations, as described in the four validation experiments of the next subsection, σ^{obs} may be assumed to be some fraction of the observation value. After consulting with Air Quality Lab personnel here at Georgia Tech, an uncertainty of 5% was applied to all the pseudo and real observations in this study.

The elements of C , the covariance matrix, are the squared estimated uncertainties in each emission source, σ^E . C constrains how much the Kalman filter will adjust E on each iteration. If C has large values, then the Kalman filter will allow large emission adjustments. Conversely, small values in C mean that the emissions are properly specified and should not be allowed to change. Equation (4) may be used to calculate a new C for each succeeding iteration. Initially, since we are uncertain about the emissions, we specify C to be equal to $50E$. However, we have found that if C is too large, C_{new} may become negative due to a numerical instability. Physically, this should not occur. To correct this problem, for the initial iteration, if C_{new} is found to be negative, C is reduced by one-half. This is repeated until C_{new} is positive, and the method is allowed to proceed.

4. Inverse Method Validation

The inverse method was validated with pseudo-observation data. Four runs were completed using the Atlanta, Georgia application of the UAM for an August 10, 1992 ozone exceedance episode. Pseudo-observations were generated by a routine UAM run (i.e. no changes from the baseline run). To test the inverse method, emissions were first perturbed from their original baseline level by multiplying the emissions field by a scalar. The model was then executed for the first hour. Next, the Kalman filter calculated an emissions adjustment that minimized the differences between the model-generated concentrations and the pseudo-observations. The inverse calculated, emissions field adjustment was added to the emissions field used previously, and the UAM was executed again for the same hour. This process was repeated until the emissions adjustment calculated in the inverse procedure was small (we chose $\Delta E < 1\%$ of $E_{original}$). The method was then repeated for succeeding hours.

We performed two pseudo-observation runs in which area source carbon monoxide (CO) emissions were increased by 40% over the original emissions used to generate the pseudo-observations. For these runs, only CO pseudo-observations at 7 surface sites were used in the inverse calculation. These 7 modeled sites (FORT, SDEK, TECH, TUCK, LOST, BRMT, and FERN in Figure 4.1) correspond to 7 real sites for which a significant amount of data exists in the SOS'1992 Atlanta Intensive data archive. Using this configuration, the problem is over-determined (i.e. 7 observations (equations) and 1 source (variable)). The inverse method will find a solution in which the differences between the model-generated concentrations and pseudo-observations are minimized. The first run uses concentration data generated from the baseline run to initialize each hour. In the second run, each succeeding hour is initialized with results from the final iteration of the previous hour. Thus in the second run, any error due to a non-perfect solution is allowed to propagate; whereas in the first run, this type of error is confined to each individual hour.

For both runs, the inverse method produced a nearly perfect solution. The small error in the solution is due to terminating the iterative process before $\Delta E=0$. Because the solutions were nearly perfect, results from the second

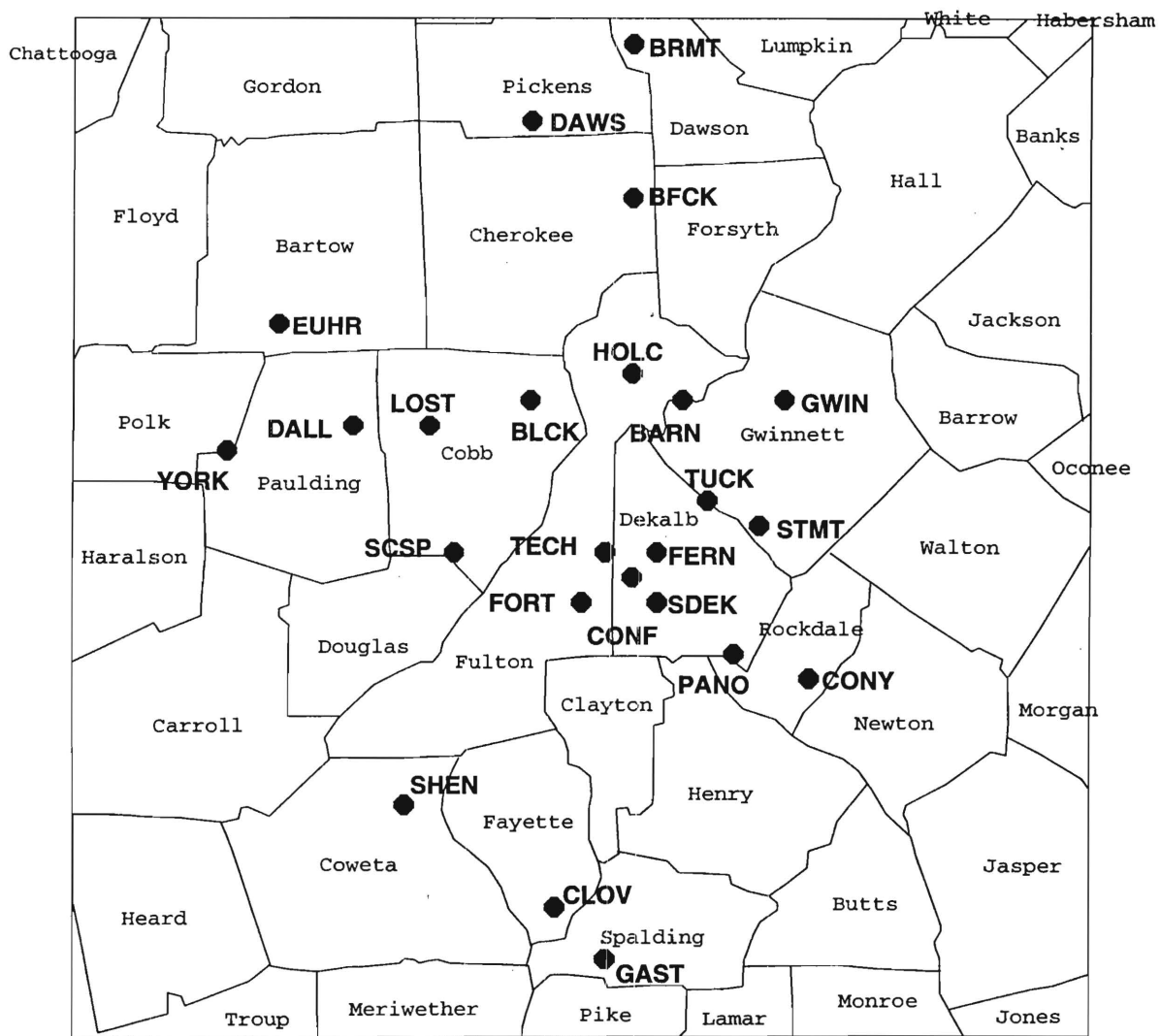


Figure 4.1. SOS 1992 Atlanta Intensive Observation Stations within the Atlanta UAM modeling domain.

run resemble results from the first; only results from the second run are shown here. Figure 4.2 illustrates the emissions adjustments calculated by the inverse method. Except for the afternoon hours, the method converged on the correct solution (-40%) after only 2 iterations. For the afternoon hours, the method required three emissions adjustments. The additional iteration in the afternoon is due to the growth of the mixing height. When the mixing height is relatively high, surface emissions are diluted into a larger volume of air than when the mixing height is low, as in the morning and late evening. As a result of the dilution, monitors at the surface are not as sensitive to perturbations in emissions. This is evident in Figure 4.3 (a) which shows the difference between the pseudo-observations of CO produced by the original emissions and the model-generated concentrations of CO produced by the perturbed (+40% area source CO) emissions. Figure 4.3 (a) also shows that the urban sites (FORT, SDEK, TECH, TUCK, and FERN) are more sensitive to changes in area source CO emissions than the rural sites (LOST and BRMT). The proximity of urban monitoring sites to area sources make them more responsive to changes in these emissions. Figure 4.3 (b) illustrates the results of adjusting the emissions by the amounts shown in Figure 4.2. At every site and for every hour, the inverse method brings the model-generated CO concentrations into agreement with the pseudo-observations.

The third and fourth runs essentially repeat the first two runs with the exception that 3 sources of CO are perturbed simultaneously. This will test the Kalman filter's ability to distinguish between different sources of air pollutants in the absence of a unique tracer for each. For both runs, area source CO emissions are increased by 60%, non-road mobile source CO emissions are increased by 30%, and mobile source CO emissions are decreased by 50% from the original emissions used to generate the pseudo-observations. The 7 observation sites utilized previously are used again in these runs.

The inverse method again performed near perfect (within the cutoff tolerance). Results from the fourth run, in which each succeeding hour is initialized with the results from the previous hour's last iteration, are shown here. Figures 4.4 (a-c) show the required emissions adjustments required by each source to bring the model-generated concentrations into closer agreement with the pseudo-observations. As before, the method efficiently finds the correct solution (-60% for area source emissions, -30% for non-road mobile source emissions, and +50% for mobile source emissions). For all 3 sources, most hours only require 2 iterations to converge on the correct solution. Observation sites that are insensitive to changes in emissions due to the growth in the mixing height or due to spatial proximity considerations may partially explain why some hours require more than two iterations to converge. However, since an emission adjustment in one source affects (feeds back on) how much the other sources should be adjusted through its impact on the observation sites, multiple iterations may simply be a consequence of the internal workings of the method. Figures 4.5 (a-b) show the before and after effects of adjusting each of the 3 emission sources on the model-generated concentrations relative to the pseudo-observations. This seems to indicate that by monitoring the concentration of one species at several sites, it should be possible to predict how much and what type of emission sources were required to produce that particular set of observations.

These experiments were presented in more detail at the *Air and Waste Management Association International Conference on the Emission Inventory: Applications and Improvements* in November, 1994. A copy of the presented paper is included in Appendix A.

5. Preliminary Control Strategy

Our original idea for developing a control strategy was to use the developed inverse method. We proposed to use the UAM run with the "base case" emissions for the specified episode and then use the Kalman Filter inverse method to determine the "optimum" emissions change needed to attain the NAAQS for ozone. Here "base case" emissions refer to the emissions of ozone precursors as determined by a combined effort of inverse method calculations (from subsection 6) and emission inventories (from subsection 1), and "optimum" refers to the least cost and most feasible emissions change.

We agreed to try to calculate a control strategy based on the emission inventories (from section 1) prior to the inverse calculations. This control strategy would have been put forth with a number of caveats since without the inverse calculations we were not yet entirely confident in the "base case" emission scenario and the ozone dependence on precursors can change dramatically depending on the precursor emissions.

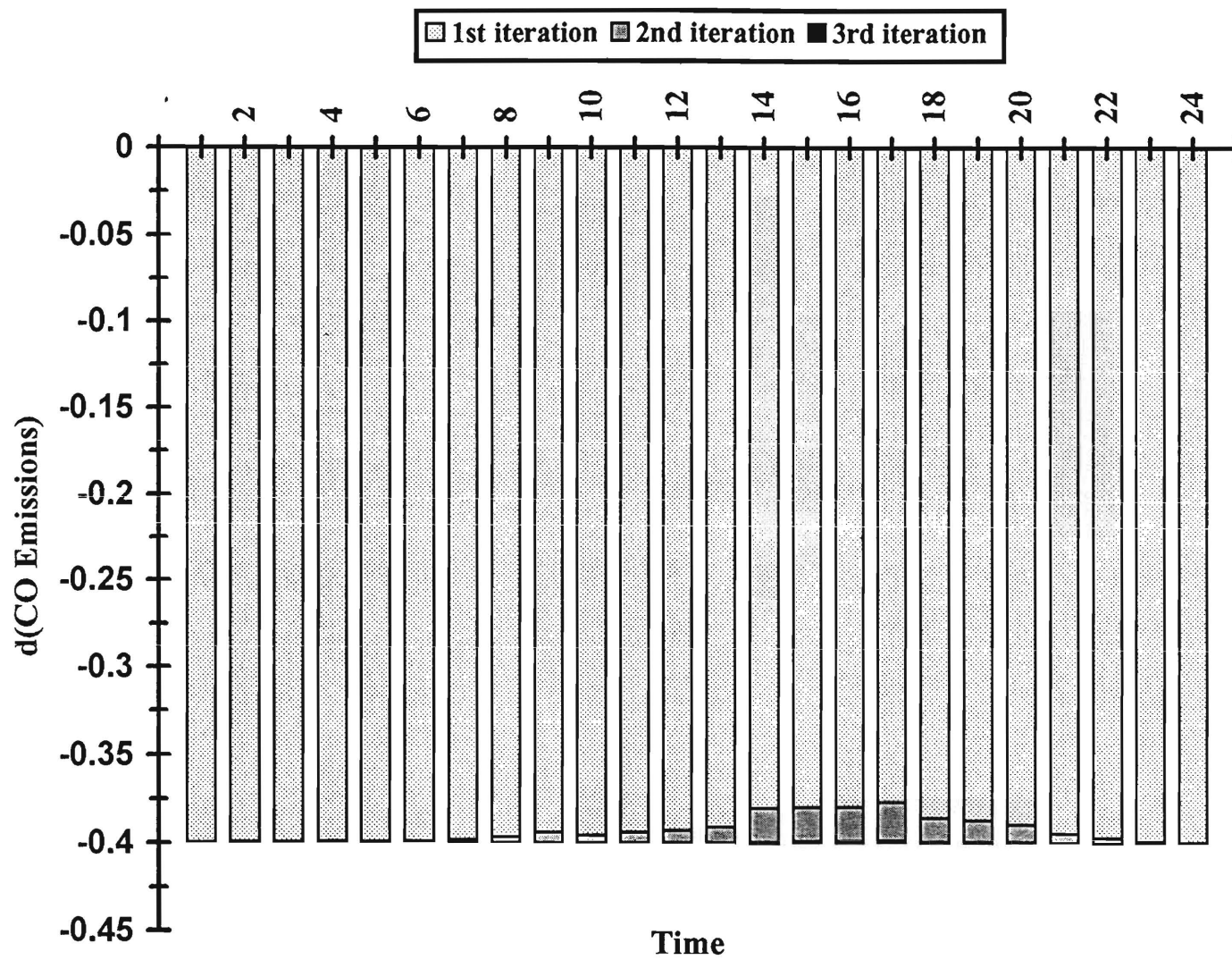


Figure 4.2. Run #2: Inverse corrections to area source CO emissions.

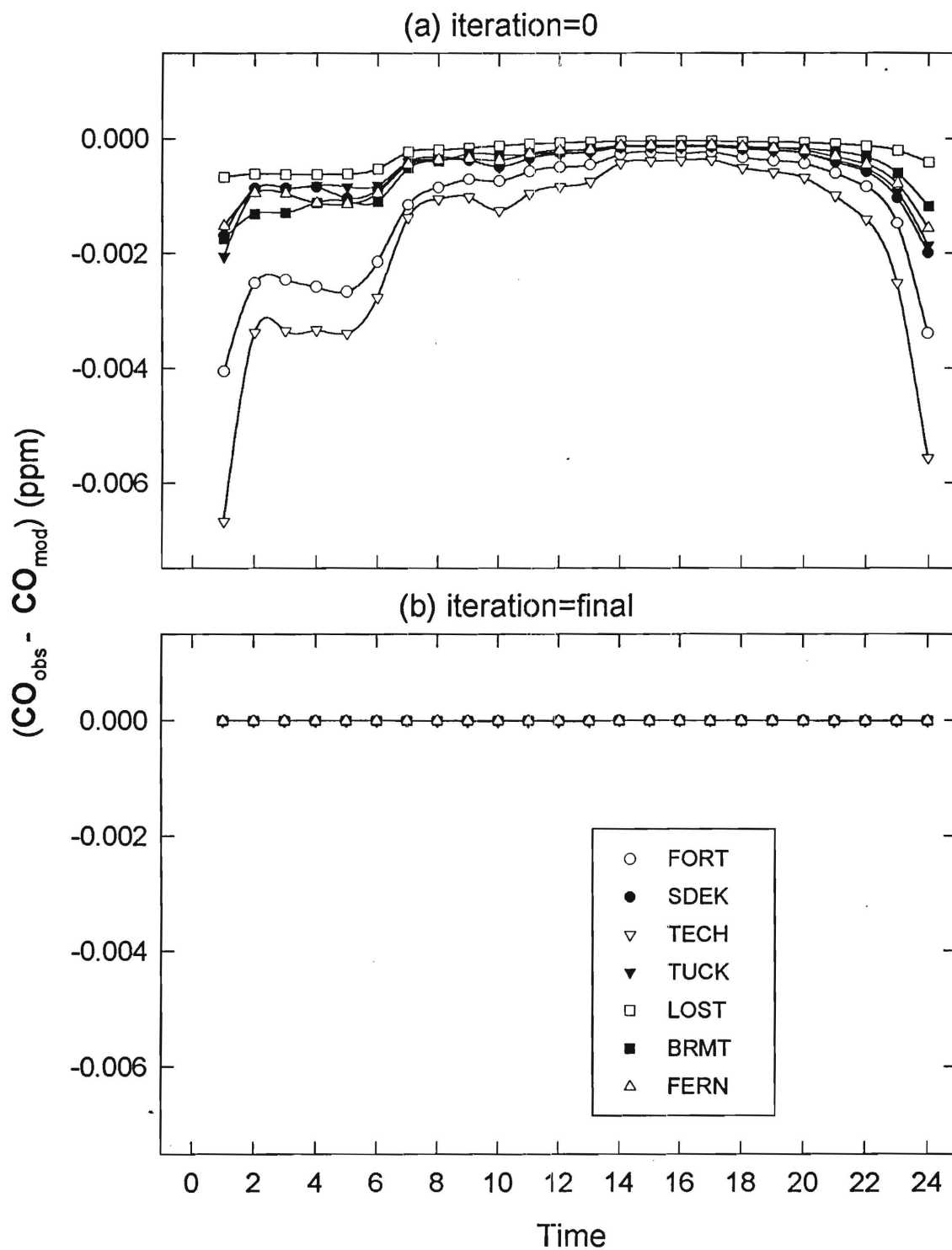


Figure 4.3. Run #2: Differences at 7 sites between pseudo-observations and model-generated CO concentrations before (a) and after (b) Kalman filter corrections to area source CO emissions.

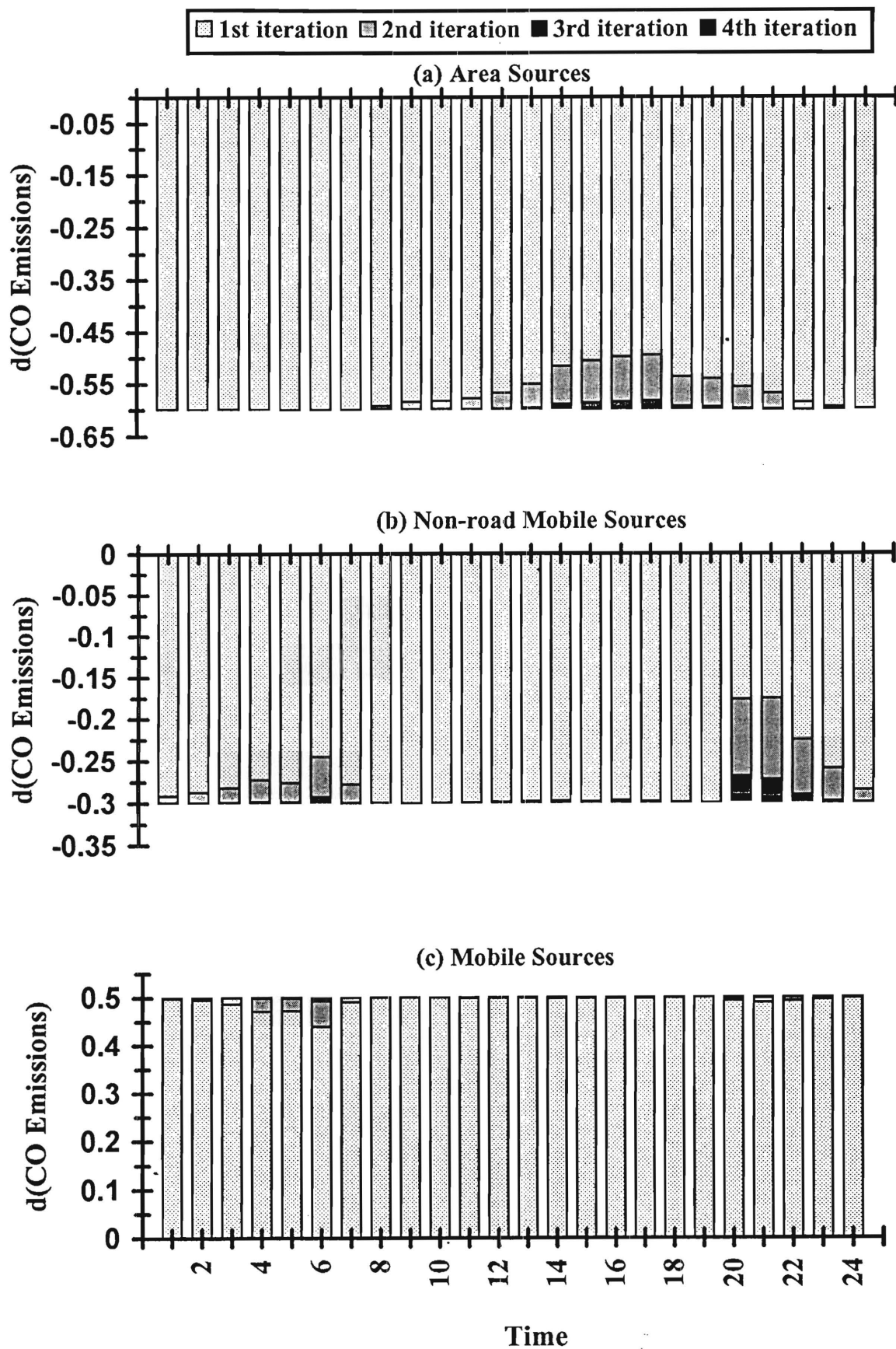


Figure 4.4. Run #4: Inverse corrections to area, non-road mobile, and mobile source CO emissions.

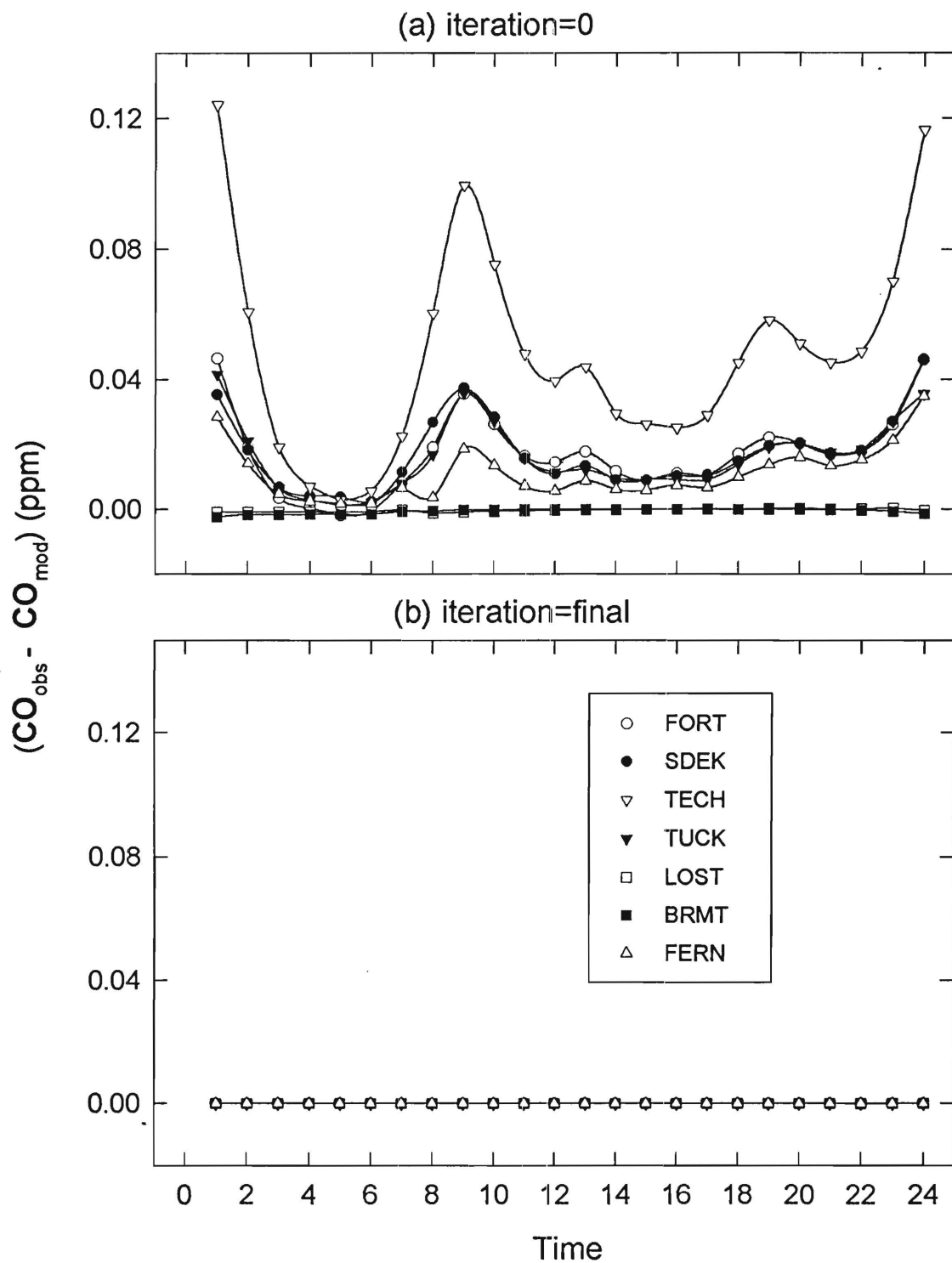


Figure 4.5. Run #4: Differences at 7 sites between pseudo-observations and model generated CO concentrations before (a) and after (b) Kalman filter corrections to area source, non-road mobile, and mobile source CO emissions.

The first step in doing this was to calculate the relationship between ozone concentrations at the different observing sites and the emissions of the various precursors. However, in order to do this, we had to assume some basic state. That is, it is too computationally demanding to calculate these relationships for all possible emissions. In Figure 5.1 we show the effect on ozone for a 10% change in (a) VOC emissions and (b) NOx emissions. Note that ozone is much more sensitive to the increase in NOx emissions than to VOC emissions. Also, the ozone is inversely related to NOx; the only way to decrease ozone at the TECH site would be to increase NOx. However, we know that there is a nonlinear relationship between ozone and NOx. In fact, we know from chemical studies that once NOx is adequately decreased the relationship will change. Since the Kalman filter assumes linearity about some state, we would have found that a dramatic increase in NOx is needed to decrease ozone. Also in figure 5.1 (c-d), we show the ratio of the results in (a) and (b) to the same calculation with a different set of emissions shown in table 5.1.

	<i>NOx</i>	<i>VOC</i>	<i>CO</i>
Point Sources	-50%	+30%	-40%
Area Sources	+50%	-40%	+40%
Non-road Mobile Sources	-40%	+40%	-30%
Mobile Sources	+40%	-50%	+30%
Biogenic Sources	-30%	+50%	-40%

Table 5.1 - Changes in Emissions from Base Case Ozone Sensitivity Runs

With these changes, we see there can be a factor of two difference in the influence on ozone. Thus, we realized that three issues were arising in our plans for a control strategy:

- ◆ we needed an optimization technique that can deal with the non-linearities
- ◆ we will therefore also need to use a more computationally demanding approach and this may require parallelization to use more efficient machines
- ◆ we needed an optimization technique that did not rely so heavily on the emissions at the time of the measurement

Thus, through this preliminary work we found that in fact our method was not adequate to address the problem.

Fortunately, there are other experts at Georgia Tech in the field of optimization. Dr. C. S. Kiang had been sitting in on some of our meetings and he arranged for us to meet with a group in Industrial and Systems Engineering (ISYE). Over the course of the last few months we have had over half a dozen meetings with them. During these meetings we have developed an approach that we plan to pursue. But we did realize that the question of developing a control strategy - and doing it correctly so that you receive information that is really useful - would be a longer process than we originally anticipated. However, as discussed above, it took some tests of our proposed method to realize its deficiencies.

Our new control strategy approach, as presented to you in a proposal this Spring, used all we have learned about the variations in the sensitivities and relationships between the gases. Thus, the effort ended up being directed at finding a new method that could work within the existing framework. It would have been detrimental to assign a control strategy with our previous method. Any control strategy would have been too full of deficiencies to be of any real use.

6. Inverse Runs using Observations from SOS 1992 Atlanta Intensive

The current regulatory model to predict biogenic emissions is the Biogenic Emissions Inventory System (BEIS). (US EPA, 1992) For a given location and meteorological conditions, BEIS calculates the gaseous chemical emissions from biogenic sources such as crops, trees, and soil. Of particular concern to air quality modelers are

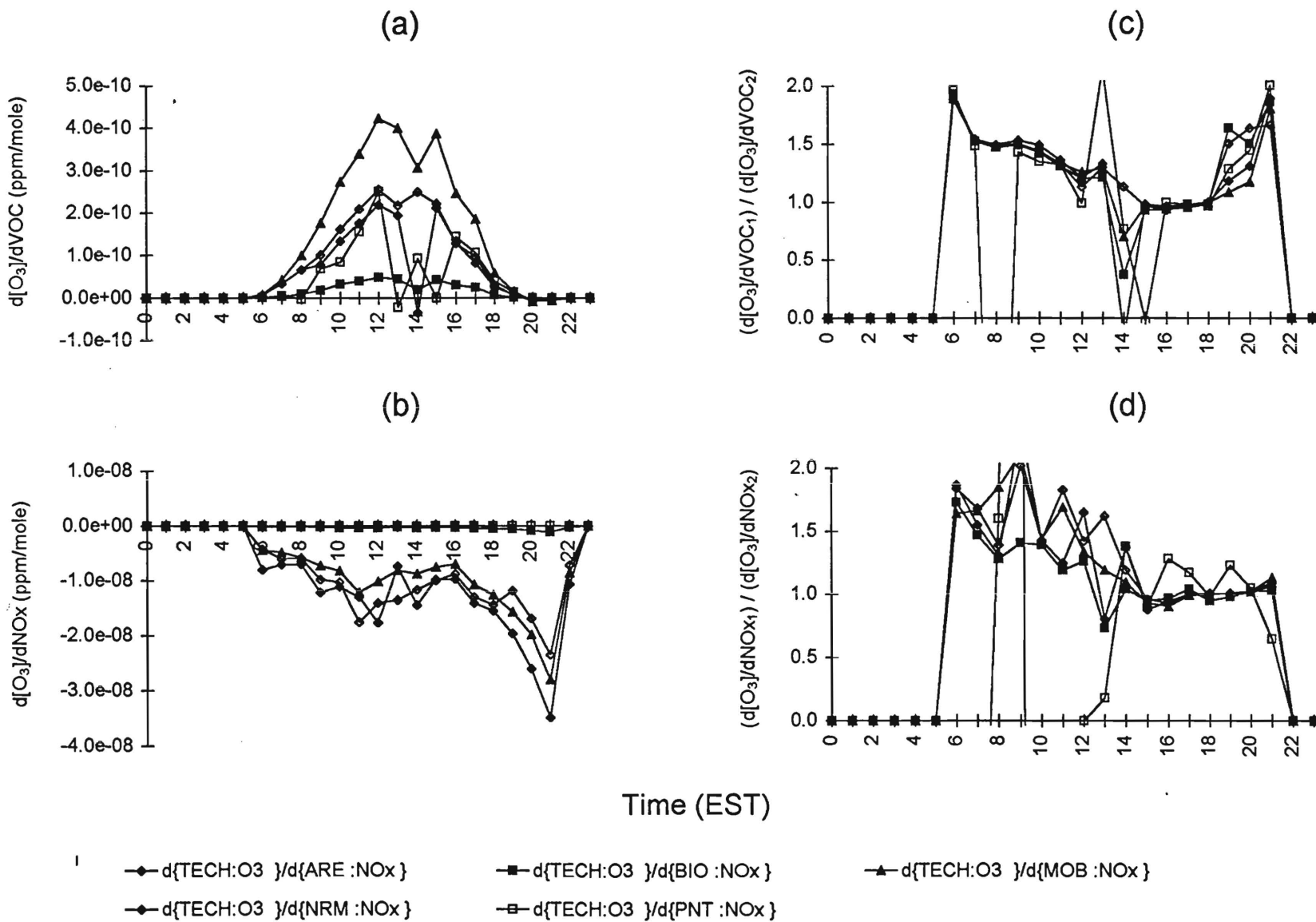


Figure 5.1. $[O_3]$ sensitivity to variations in VOC and NO_x Emissions at Georgia Tech.

emissions of isoprene. Isoprene is a highly reactive ozone precursor of primarily biogenic origin. Relative to cities in other regions, southeastern cities, including Atlanta, have a large number of trees in and around the urban core. Thus, in southeastern cities, isoprene may play a potentially large role in ozone formation.

Figure 4.1 showed the locations of the observation stations in the Atlanta UAM domain during the 1992 SOS Atlanta Intensive. Included as appendix C are preliminary observed chemical data (inorganics and organics), averaged over one hour, and speciated into the Carbon Bond - IV mechanism for direct comparison with UAM output. For isoprene, there were 5 continuous monitors (Fort McPherson, Lost Mountain, South Dekalb, Georgia Tech, and Tucker) operating on August 10. Further, there were other sporadic can samples taken throughout the day at various locations. All these data were used to compare and reconcile the differences between observed and predicted isoprene concentrations.

Using BEIS, biogenic sources account for 76% of the total hydrocarbon emissions for the August 9-10, 1992 Atlanta UAM modeling inventory. Despite their prominence in the emission inventory, figure 6.1 (a) shows that the UAM underpredicts airborne concentrations of isoprene by 85%¹ when compared to observations for August 10, 1992.

In March 1995, the EPA suggested regulatory agencies use a new version of the Biogenic Emissions Inventory System (BEIS2). (Hunt, 1995) BEIS2 updates the emission factors, land use inventories, and environmental correction factors used to calculate biogenic hydrocarbon and NO_x emissions. For a two week summer 1988 simulation with the Regional Oxidant Model (ROM), the EPA reported a five-fold increase in isoprene emissions from BEIS2 relative to BEIS. As shown in figure 6.2 (a) and (b) for the August 10, 1992 Atlanta inventory, the spatially averaged, BEIS2 isoprene emissions were 3 to 5 times greater than BEIS emissions. Averaged over the 12 hour simulation, BEIS2 emissions were ~4 times higher than BEIS emissions. However, figure 6.1 (b) shows that with BEIS2, the UAM still underpredicts isoprene concentrations by 34%¹.

Initializing with emissions from BEIS, figure 6.1 (c) shows that the inverse method brought predicted isoprene concentrations within -8%¹ of observed isoprene concentrations. To achieve this agreement, figure 6.2 shows the isoprene emissions were increased for some hours (as much as a factor of 12 at t=13), and decreased for other hours (as small as 0 for hours 16, 17, and 18). Over the 12 hour simulation, the average adjustment was a factor of ~5.

Initializing with emissions from BEIS2, figure 6.1 (d) indicates that the inverse method improved the prediction of isoprene concentrations to within -9%¹ when compared to observations. While not needing as much adjustment as BEIS, figure 6.2 shows that BEIS2 emissions may still be underestimated as much as a factor of ~3.

Relative to BEIS, BEIS2 improves the temporal distribution and magnitude of emissions. However, it does not improve the spatial allocation of those emissions. Figure 6.3 (a) and (c) shows the total daily biogenic isoprene emissions calculated by BEIS2 and the inverse corrected BEIS2 calculation. Figure 6.3 (b) is the ratio of BEIS2 to the corrected BEIS2. Over the whole day, the corrected BEIS2 emissions are about 35% higher than BEIS2. From this graphic it is evident that BEIS2 only resolves emissions down to the county level. For example, downtown Atlanta in Central Fulton County has the same amount of emissions as the more densely forested areas of North and South Fulton County. To investigate this deficiency, we split the biogenic emissions into rural and urban components. Using data from the EPS2 surrogate file, the percentage of urban land area was compared to the percentage of rural land area. The grid cell was characterized as urban if the urban coverage was greater than the rural coverage and vice versa. A filter was then programmed to separate the biogenic emissions into the urban and rural components based on the grid cell classification.

Now that urban and rural biogenic emissions could react independently, the inverse method was applied again. When compared to observations, figure 6.1 (e) shows the method improved the prediction of isoprene

$$^1\text{normalized bias} = \frac{\sum_{i=1}^N (\chi_i^{obs} - \chi_i^{pred}) / \chi_i^{obs}}{N}; \text{calculated for the 5 continuous monitors for hours 7-17.}$$

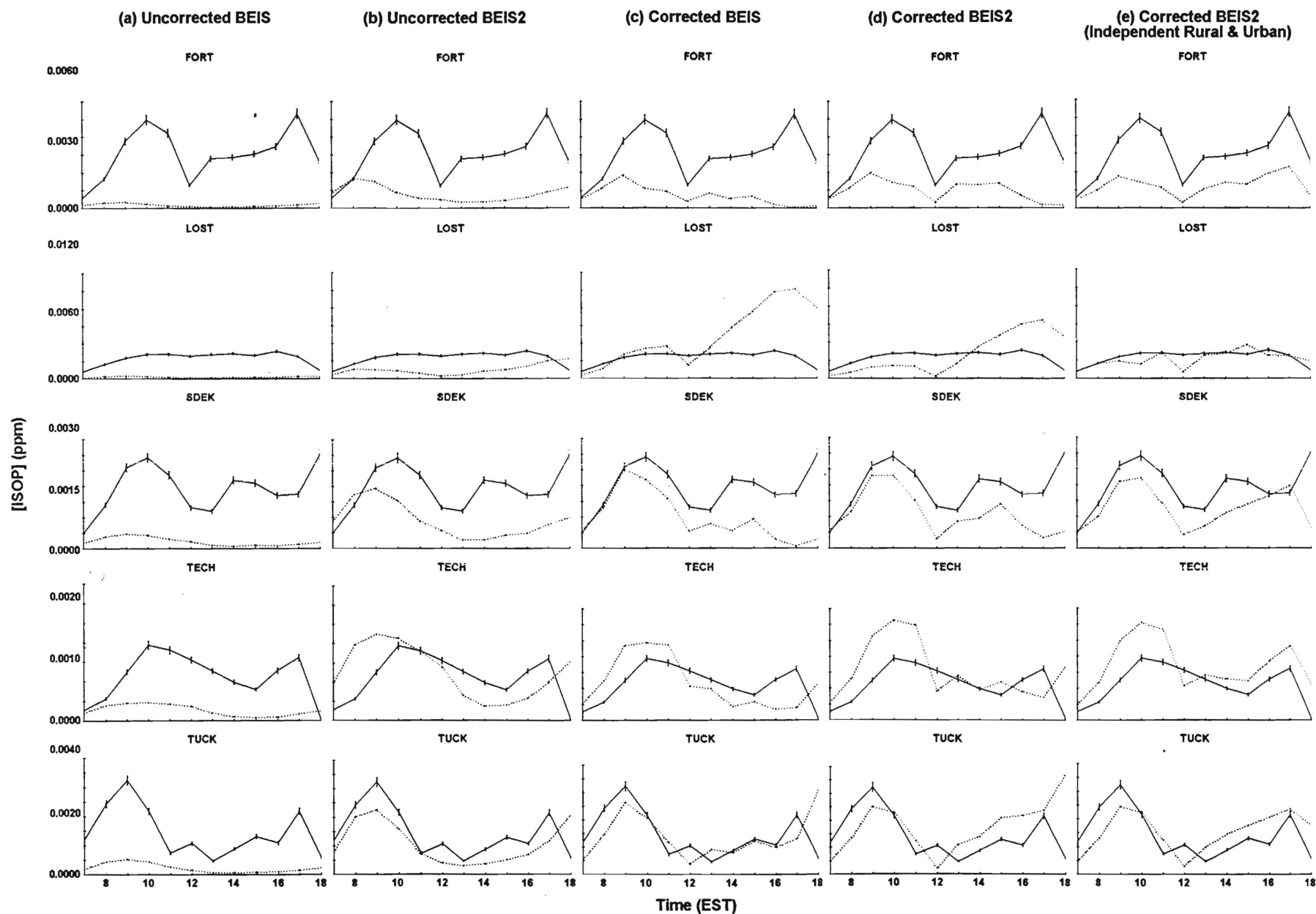


Figure 6.1. Observed (solid line) and UAM predicted (dashed line) concentrations of isoprene with and without inverse calculated corrections.

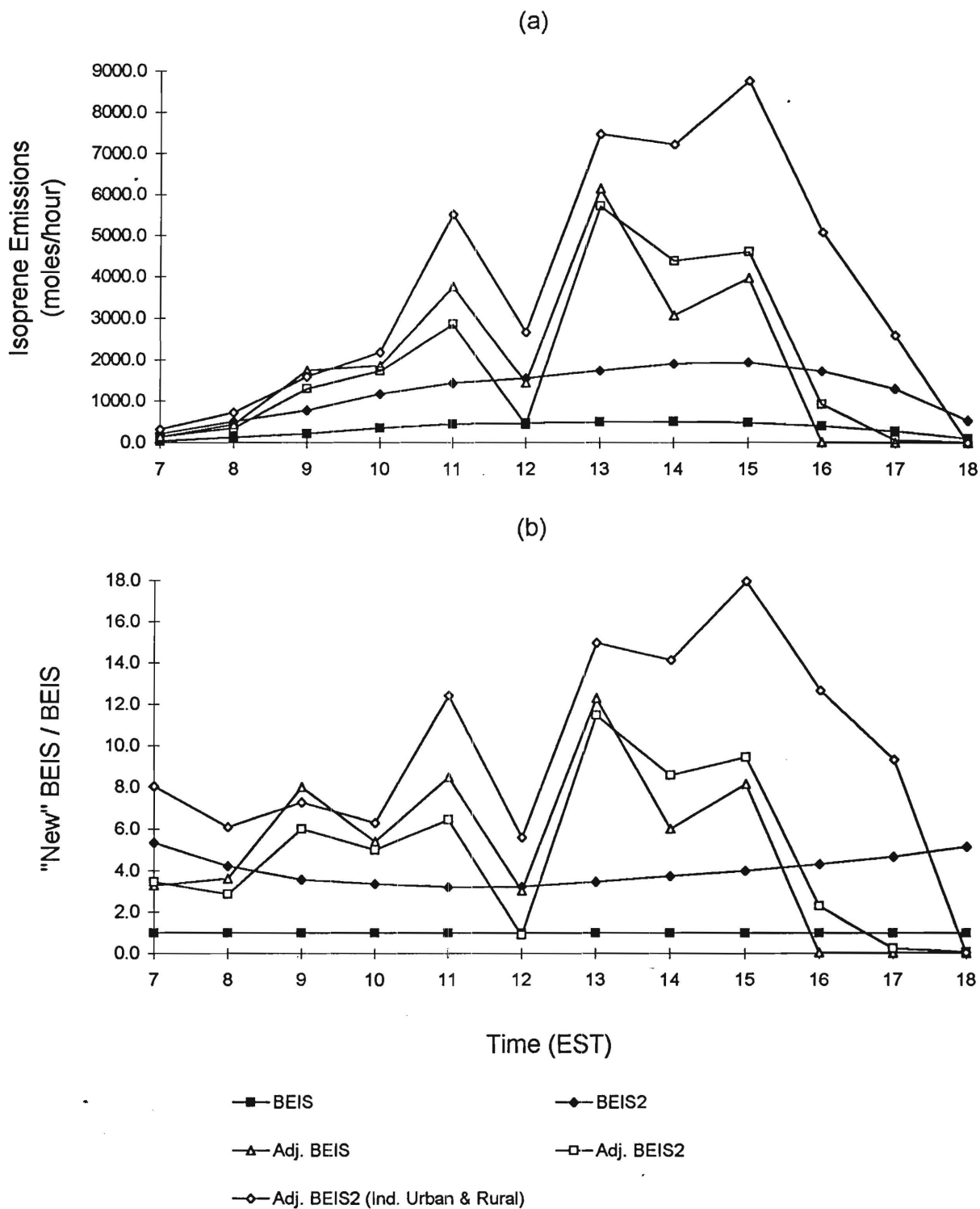


Figure 6.2. (a) Average isoprene emissions. (b) Ratio of average isoprene emissions to BEIS.

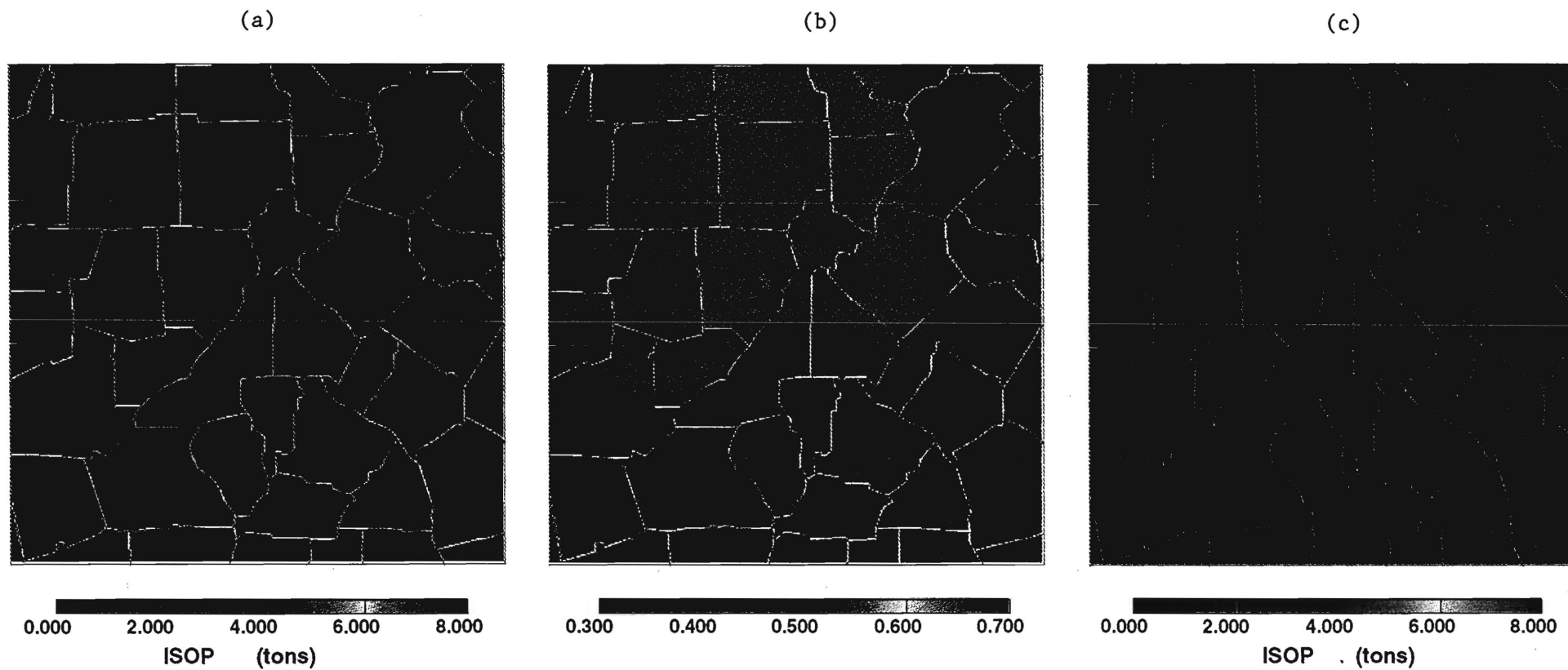
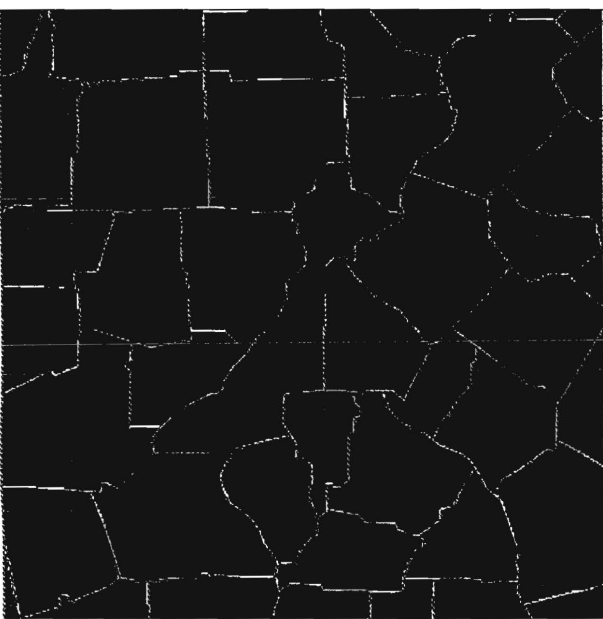


Figure 6.3. Daily BEIS2 (a) and inverse adjusted BEIS2 (c) isoprene emissions. Ratio (b) of (a) to (c).

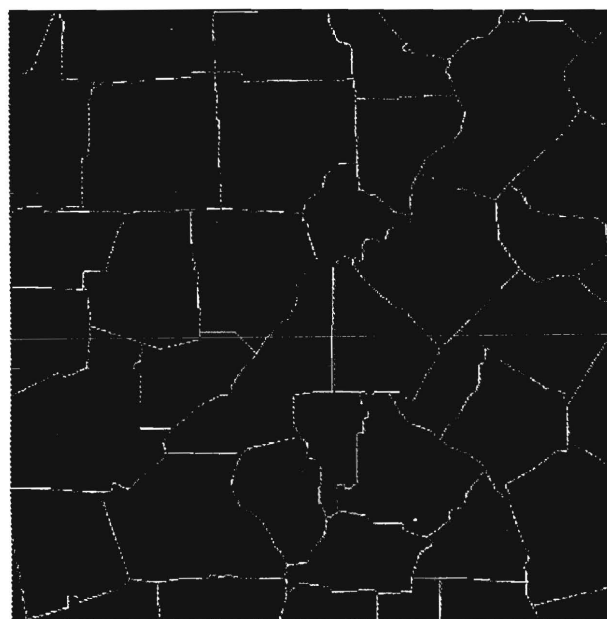
(a)

(b)

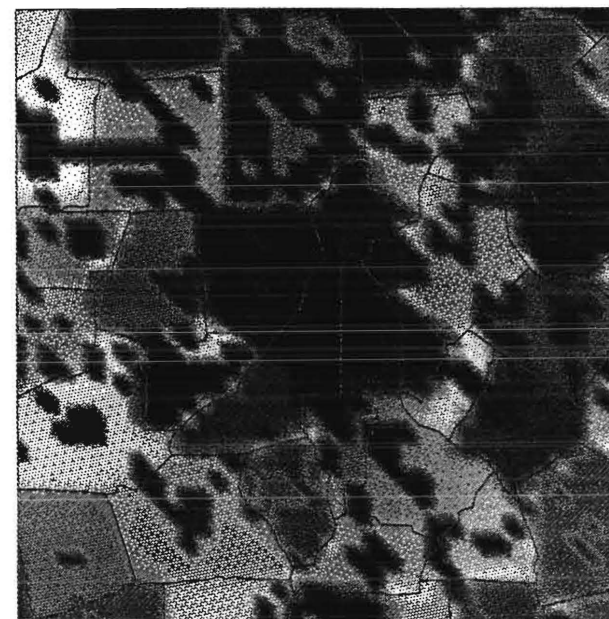
(c)



0.000 2.000 4.000 6.000 8.000
ISOP (tons)



0.300 0.400 0.500 0.600 0.700



0.000 2.000 4.000 6.000 8.000
ISOP (tons)

Figure 6.4. Daily BEIS2 (a) and inverse adjusted BEIS2 with independent urban and rural isoprene emissions. Ratio (b) of (a) to (c).

concentrations to within -5%¹. Figure 6.2 shows the spatially averaged temporal change in isoprene emissions. The total daily isoprene emissions for BEIS2 and the corrected BEIS2 with independent acting urban and rural components are shown in figure 6.4 (a) and (c). Figure 6.4 (b) is the ratio of BEIS2 to the adjusted BEIS2 with independent urban and rural components. Relative to BEIS2, figure 6.4 shows that the inverse method adjusted the biogenic isoprene emissions by a factor of ~3 in the rural areas, but only by a factor of ~2 in the urban areas. Also note in figure 6.4 (c) that the urban areas now have less biogenic emissions relative to the rural areas, as we would expect. Using the UAM, this final biogenic isoprene inventory most closely matches what was observed in the atmosphere in 1992.

7. Activity Factor / Emission Factor Improvement

a. Biogenic Sources

A new procedure for estimating biogenic emissions was created by Thomas Pierce and others from the EPA. The new method abandons the traditional tree classification of deciduous, coniferous, and mixed forests. Instead, the method emphasizes emissions coming from each specific variety of tree present in an area. Our group at Georgia Tech, working jointly with Mr. Pierce, was able to generate a computer code, named BEIS2, that incorporates the new algorithm into EPS2. This new code is available to the GA DNR.

We performed several numerical computations using BEIS2 to explore the impact of the new biogenic estimates on ozone concentrations and on control strategies. All the UAM simulations described in this section are for August 9 and 10, 1992. The results of these computations can be summarized in the following way:

- ◆ The biogenic emission contribution to the total VOC within the Atlanta UAM domain changed from 55% (29% isoprene and 26% monoterpenes) using BEIS, to 76% (59% isoprene and 17% monoterpenes) using BEIS2.
- ◆ The maximum ozone concentration changed from 129 ppbv with BEIS, to 169 ppbv with BEIS2. The total ozone production increased by 14% with the new biogenic emissions.
- ◆ Using 35% reductions in anthropogenic NO_x and VOC, the Atlanta region changed from being VOC-limited under BEIS, to NO_x-limited under BEIS2.

In figure 7.1, the change in UAM predicted O₃ at all 1600 surface grid cells due to a 35% reduction in anthropogenic (a) NO_x and (b) VOC emissions with biogenic emissions from BEIS is shown as a function of the basecase daily maximum O₃ concentration. For a significant number of grid cells, reducing NO_x increases O₃ (negative reductions). VOC reductions either have no effect or always reduce O₃ concentrations. Figure 7.1 (c) and (d) shows the same as (a) and (b) except that biogenic emissions were calculated using BEIS2. Using BEIS2, there are no significant increases in O₃ when NO_x is reduced. Further, contrary to what we expected, reductions in anthropogenic VOC emissions were more effective in reducing O₃ across the domain despite the increase in biogenic hydrocarbons.

b. Area Sources - Auto Refinishing and Graphic Arts

As part of the 1992 Atlanta Intensive Study a survey on emissions and activity level was conducted for two area sources: automobile refinishing and graphic arts. We calculated the annual average emissions from each automobile refinishing facility based on the amount of solvent used multiplied by the corresponding emission factor obtained from the EPA document, AP-42. For graphic arts, the VOC emissions were calculated by multiplying the amount of material consumed, by the corresponding emission factor which was derived from the physical properties of the material used at each facility. To obtain the VOC emissions at the county level, we developed an alternate factor that converts the emissions obtained from the graphic arts facilities that answered the surveys, within a given county to the county total VOC emissions. This alternate factor was calculated by taking the employment data listed in the County Business Patterns divided by the number of employees for each county. This procedure allowed us to compare our emission factor with the EPA's per employee factor. The results are summarized as follows:

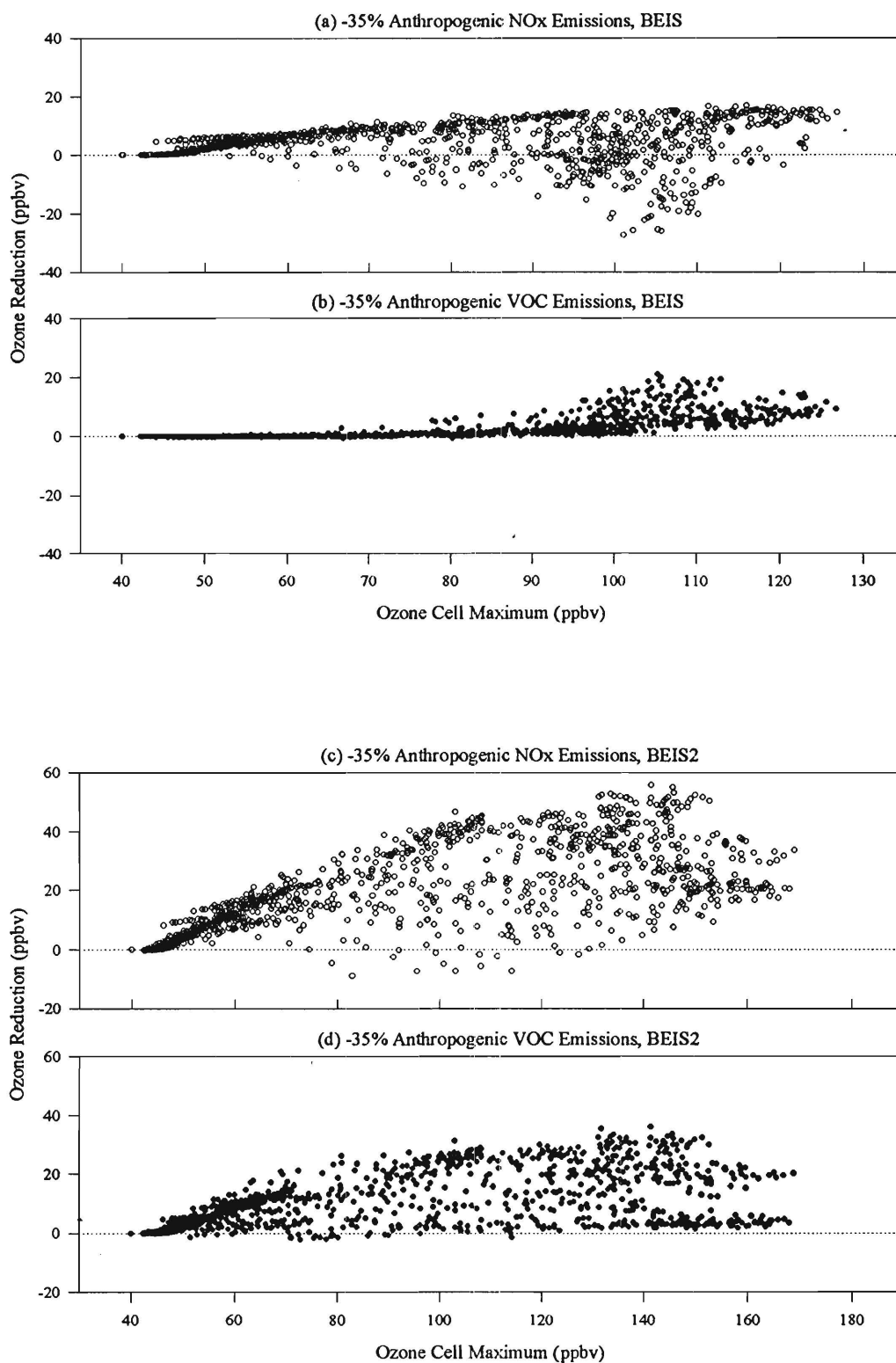


Figure 7.1 Ozone sensitivity to anthropogenic No_x and VOC reductions using BEIS (a-b) and BEIS2 (c-d).

- ◆ For automobile refinishing, the regression analysis suggested that there is about a factor of two between the EPA's per employee factor (3,519 lbs/employee/year) and the factor derived from the surveys (1,656 lbs/employee/year) ($R=0.98$).
- ◆ For graphic arts, the emissions from Fulton County appear to be different than those from the other counties. The regression analysis suggests that our emission factor is in agreement with the EPA's per capita factor when excluding Fulton County ($R=0.90$).
- ◆ For both automobile refinishing and graphic arts, our analysis of the daily activity logs suggests that the 5 days/week operating schedule used in the current emission inventory is a valid assumption.

These findings were presented in more detail at the *Air and Waste Management Association International Conference on the Emission Inventory: Applications and Improvements* in November, 1994. A copy of the presented paper is included in Appendix B.

8. Other Research

a. Point Source Emissions - Impact on Control Strategies

Within the Atlanta UAM domain, the point sources represent about half of the total anthropogenic NO_x emissions. About 90% of point source NO_x emissions were from four Georgia Power Plants: Bowen, McDonough, Wansley, and Yates. Each comprises about 43, 7, 26, and 14 % of the point source NO_x emissions. Thus these sources are potentially important when developing control strategies. In evaluating the impact of the NO_x emissions from these power plants on the O₃ chemistry within the modeling domain, we found the effect varies depending on the location of the power plant (ie. a rural or urban). Four UAM simulations were made: base case, base case without Bowen, base case without McDonough, and base case without both Bowen and McDonough. Removing the emissions from McDonough results in an O₃ increase while removing the emissions from Bowen results in an O₃ reduction at the region downwind from that power plant. Plant Bowen is located in a rural area, about 60 km northwest from downtown Atlanta, where it is the main source of NO_x emissions. On the other hand, McDonough is located in an urban area where NO_x is abundant and an excess of NO_x actually scavenges O₃. The maximum change in O₃ concentration is up to -53 ppbv and +38 ppbv, respectively. Furthermore, the area affected by Bowen is much larger than McDonough because it contributes nearly half of the point source NO_x emissions. Although removing the emissions from Bowen can result in an O₃ reduction, the O₃ concentration in most of the affected region was already in attainment. The UAM simulation for the base case without both Bowen and McDonough indicates that the two plumes are acting independently (ie. the plumes are not mixing together within the UAM domain).

These findings were presented in more detail at the *Air and Waste Management Association International Conference on the Emission Inventory: Applications and Improvements* in November, 1994. A copy of the presented paper is included in Appendix B.

b. Ozone Sensitivity to Meteorology

It is well known that meteorological parameters are critical to the UAM ozone simulations. We explore the sensitivity of UAM simulations to meteorological variables in two different situations. In the first case, we compared the BEIS2 simulation (identified as the Tech simulation) with an SAI-generated simulation. Both simulations are identical with respect to emission inventories but differ in the wind fields and in the boundary conditions. The results showed that:

- ◆ The maximum ozone concentration changed from 169 ppbv if the Tech simulation was used, to 201 ppbv if the SAI simulation was used; the total ozone production increased by 14% with the SAI meteorology.
- ◆ In the second case, the Tech simulation was compared with a lidar-generated mixing height simulation in which the only change was that the mixing height profile was derived from lidar observations. The results showed that the maximum ozone concentration changed from 169 ppbv if the Tech simulation was used, to 131 ppbv if the lidar-generated mixing height was used. The total ozone production decreased by 3% with the lidar measurements.

These results demonstrate the considerable uncertainty in UAM calculations due to meteorological factors.

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III. Appendices

Appendix A - Papers

Appendix B - Code

Appendix C - Data

G-35-803
#20 (New)


Inverse Modeling for Atlanta, Georgia

Final Progress Report (GTRC contract #771-390279)

Submitted to
Air Protection Branch
Environmental Protection Division
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July 1, 1996

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I. Executive Summary

In our proposal of April 25, 1995, we planned to continue the inverse modeling project to resolve the Urban Airshed Model (UAM) predictions of isoprene, carbon monoxide (CO), volatile organic compounds (VOCs), and nitrogen oxides (NO_x) with observations taken during the 1992 Southern Oxidants Study (SOS) Atlanta Intensive. Isoprene has been completed. Due to an earlier error in our inverse code, previous findings regarding isoprene are superseded by the results presented here. These revised results have been submitted for peer review and publication in *Geophysical Research Letters*. Using pseudo-data, we were able to successfully run one inverse for CO and one for NO. However using real data, we have not been able to use the inverse method to resolve the emissions of either of these species. We believe that the difficulty lies in the spatial distribution of the emission sources. Whereas isoprene sources (all biogenic) are numerous, small, and ubiquitous, sources of CO and NO (and anthropogenic sources of VOCs too) are large, relatively concentrated, and highly spatially dependent. A small error in the spatial allocation of these sources in the emission inventory may cause a large difference between model predicted ambient concentrations and observations at any one point in space and time. Because the Kalman filter is driven by the differences between observations and model predictions, these small errors are translated into large fluctuations in the emissions. The resultant emissions reflect the errors in the spatial allocation of the inventory rather than the magnitude of the emissions. Unfortunately, there is not enough observation data to "unlock" the spatial constraints and use the inverse method to solve for the magnitude *and* spatial distribution of the anthropogenic emissions.

The remainder of this report describes the updated isoprene results and documents the CO and NO_x inverse attempts. Anthropogenic volatile organic compounds share many of the same characteristics as CO and NO_x. Thus, due to the problems associated with CO and NO_x, we did not attempt to run an inverse with VOCs.

II. Inverse Modeling for Atlanta, Georgia

1. Isoprene - Revised Results

Since our last presentation, we discovered coding errors in our inverse application. These errors were in the initialization of isoprene at each hour and in some of the details of the error matrices. The code corrections resulted in emission correction factors that differed from the values we originally reported, however the conclusions are the same. The table below summarizes the original values and the revised values.

Inverse Derived Relative Scaling Factors w.r.t. BEIS or BEIS2

	BEIS	BEIS2	Split-BEIS		Split-BEIS2	
			urban	rural	urban	rural
Original	18	4	17	20	3	5
Revised	7	2	10	6	2	2+

Due to the correction, negative emissions occur at various times throughout the day as shown in figure 1. At hour 7, the inverse method adjusted the emissions more to compensate for errors in the initial conditions than for errors in the emission inventory. The physical implication of the negative emissions at hours 11, 12, 16, 17 and 18 indicate that the model could be lacking a key isoprene removal mechanism, or that the inverse method is correcting for an overestimation of earlier emission adjustments. For hours 16 and 17, the model assumes clear sky although rain was recorded in Atlanta. The impact of this assumption on comparisons between observations and model predictions is not clear. Because of this uncertainty, the data are omitted from the isoprene emission inventory comparisons when rain may have interfered (hours 16, 17, and 18). No physical explanation corresponding to the negative emissions at hours 11 and 12 has been found. These emissions cannot be omitted because it is uncertain whether an unknown removal mechanism acted at these hours, or if the method is correcting itself for an over estimation of isoprene emissions during the morning hours (8, 9, and 10).

In addition to the change in the magnitude of emissions, results from the split-inventory runs show that the spatial allocation of emissions is also different than the distribution originally reported. Emissions in the urban areas are higher than previously shown. This finding is in better agreement with the latest emissions research (Geron et al. 1995).

The code changes and different emissions results also affected the model performance statistics. The table below shows the original and revised values.

Model Performance (Normalized Bias) relative to Observations

	BEIS	BEIS2	Inverse-adjusted BEIS	Inverse-adjusted BEIS2	Inverse-adjusted Split BEIS	Inverse-adjusted Split BEIS2
Original	-85%	-30%	+3%	+7%	+1%	+2%
Revised	-83%	-19%	-15%	-9%	-8%	-5%

The change in emissions did not affect the ozone simulation. Although the increase in emissions is smaller than originally reported, they are still sufficient to saturate the region with hydrocarbons. Additional isoprene (as in the original run) has no effect on the ozone beyond the stoichiometric level.

All of the above results are described in detail in the Appendix: a copy of our revised paper "Inverse Modeling of Biogenic Isoprene Emissions" submitted to *Geophysical Research Letters* on 6/15/96.

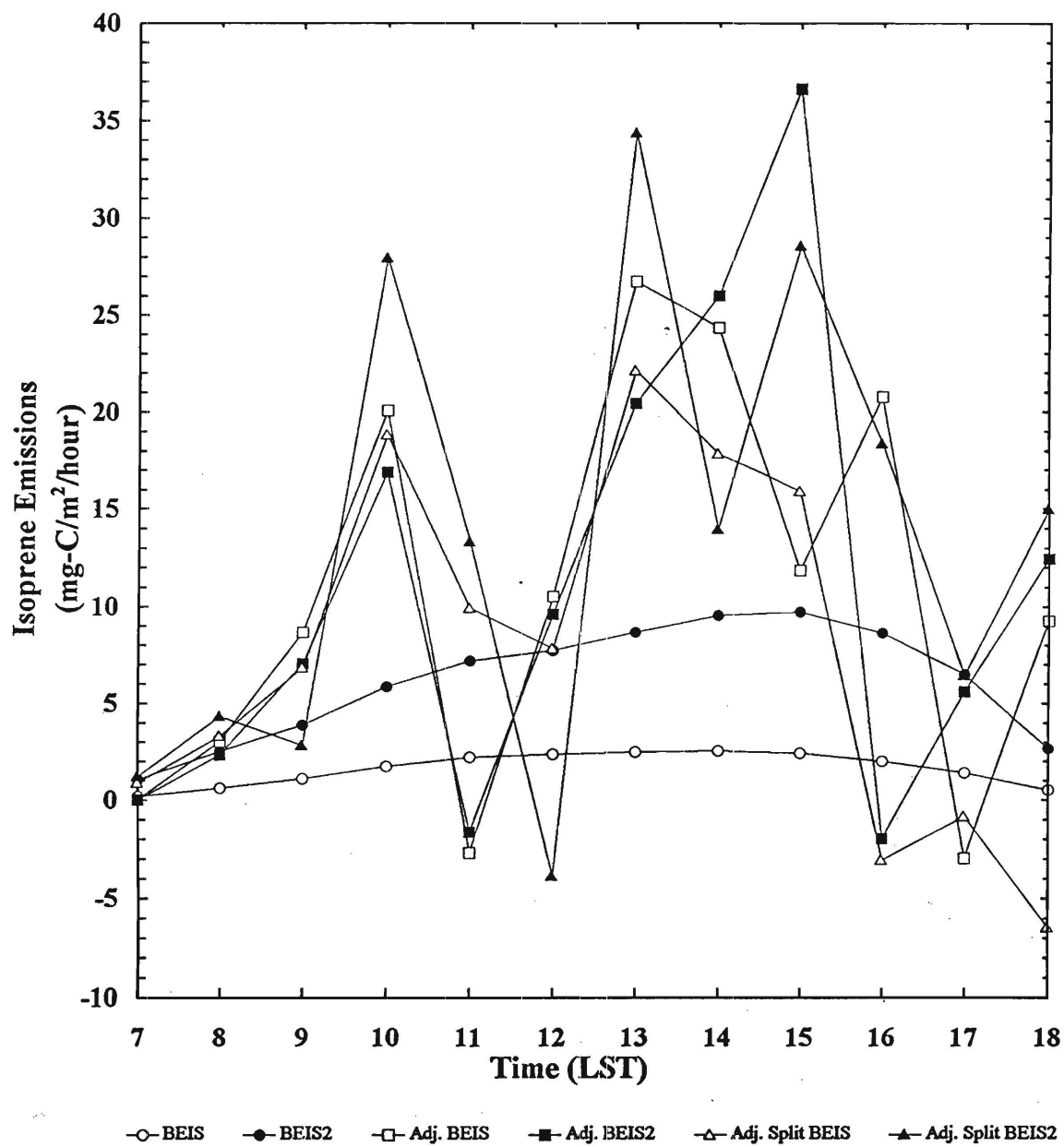


Figure 1. Revised inverse derived average isoprene emissions for Atlanta, Georgia (August 10, 1992).

2. Carbon Monoxide (CO)

On August 10, 1992, there were four (4) carbon monoxide (CO) monitors operating in the Atlanta Area. The Georgia Tech site (TECH) was the only true urban site. Both South Dekalb (SDEK) and the Holcomb Bridge Road (HOLC) sites may be considered as suburban. Finally the Yorkville site (YORK) was the only rural station. Figure 2 shows the observed concentrations and initial UAM predicted (prior to any adjustments) concentrations. At the TECH site, CO concentrations are overpredicted by 270%; at the SDEK site, the UAM overpredicts CO by 6%; at HOLC, the model underpredicts CO by 41%; and at YORK, CO concentrations are overpredicted by 15%. In total, the UAM overpredicts CO concentrations by 64% (mostly due to the large error at TECH).

A. CO Inverse with Pseudo-data

A pseudo-data CO inverse run was used to determine the optimum settings for the Kalman filter. Area, nonroad mobile, and mobile sources were all merged into one gridded and temporally allocated source. Point sources were ignored since their contribution to the CO inventory was minor. This single source was then initially set to 160% of the baseline. The filter was then used to determine the base emissions using only the differences between the predicted CO concentrations (with initial emissions at 160% of the baseline) and the observed concentrations (pseudo-data - CO concentrations extracted from a baseline run). Figure 3 shows the pseudo-data observations and the initially perturbed concentrations at each of the 4 sites. Figure 4 shows the inverse derived factors necessary to force the model to match the pseudo-data. In this case, a value of 1.00 implies a perfect convergence. Deviations from 1.00 are the percent error in emissions from true convergence. Thus, for a 60% error in the emissions, we concluded from this pseudo-data run that the inverse method is capable of resolving a single domain-wide CO source.

B. CO Inverse with Observations from the 1992 Atlanta Intensive

Using observations of CO from the 1992 Atlanta Intensive (see figure 2) and the August 10, 1992 (day specific) CO emission inventory, an inverse was run to estimate the temporal distribution of CO emissions assuming that the spatial distribution of the emissions is correct. Figure 5 shows that the inverse was successful in forcing the UAM predictions to match the observations. Overall, the bias was reduced from +64% to -18%. In particular, the adjusted emissions vastly improved the simulation for the TECH site (at the expense of the SDEK site). The biases at the TECH, SDEK, HOLC, and YORK sites were +16%, -43%, -52%, and +8% respectively.

The inverse-derived CO emission adjustment factors are shown in figure 6. It is clear that the oscillations in the emissions are unrealistic. In an attempt to dampen these oscillations, we adjusted several filter input parameters and ran the inverse again. In this new run, the emissions covariance was decreased from 50 to 0.2. This parameter is a measure of the uncertainty in the emissions inventory (a large value indicates a large uncertainty and a willingness to allow the emissions to fluctuate freely to ensure the best fit between observations and model predictions; a small value indicates confidence in the emissions inventory and allows only small "fine-tune" emissions adjustments). The threshold for convergence was also increased to 5% from 1%. The method finds the best solution by iteration. It stops when the change in emissions is less than the threshold value. Increasing the threshold limits the amount that the final solution may differ from the initial guess by stopping the iterative process early (and perhaps prematurely). By "tuning" these parameters, a satisfactory and stable solution is obtained. Overall at all stations, the bias is reduced to just +3%. The biases at the TECH, SDEK, HOLC, and YORK sites were +73%, -24%, -47%, and +12% respectively (see figure 7). Further, the emissions seem to be more well behaved. Figure 8 shows that the CO inventory is overestimating CO emissions in the morning and late evening hours and is about correct during the daylight hours.

Originally, the parameters that were changed in the last run were initially set to optimize the performance of the pseudo-data run. They were then changed to optimize the performance of the real-data run. To determine if the run with the new parameters is a legitimate solution, the pseudo-data test was run again with the new parameters. Again, the emissions were perturbed to 160% of the baseline value. Rather than the relatively perfect solution seen in the initial pseudo-data run (see figure 4), figure 9 shows that the solution begins to degrade at hour 8 and could

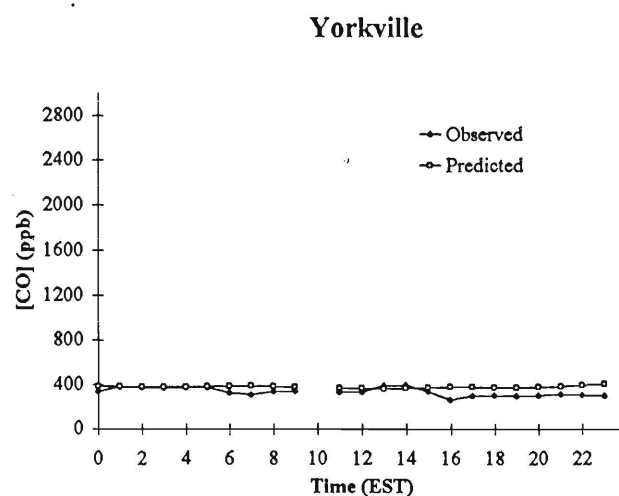
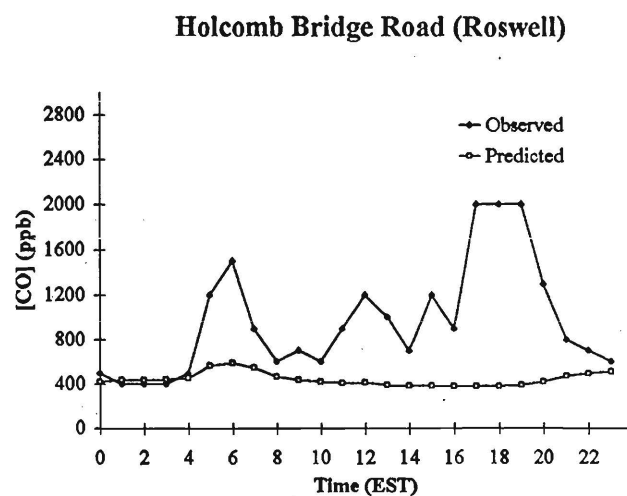
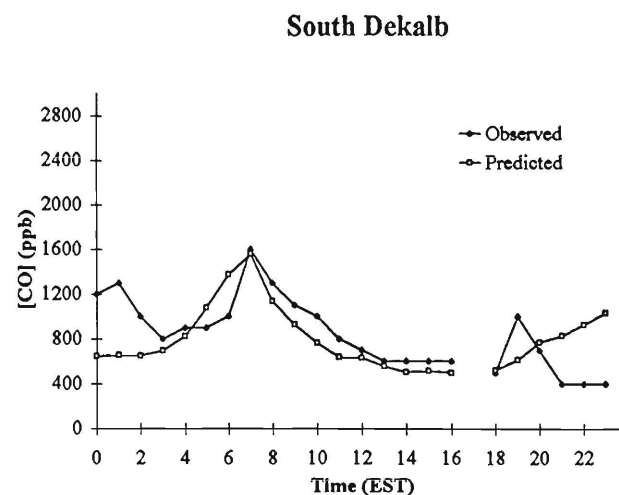
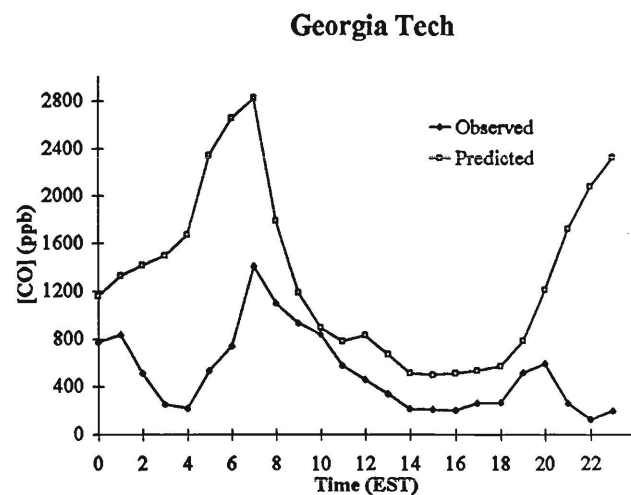


Figure 2. Observed and initial UAM predicted CO concentrations in Atlanta, Georgia (August 10, 1992).

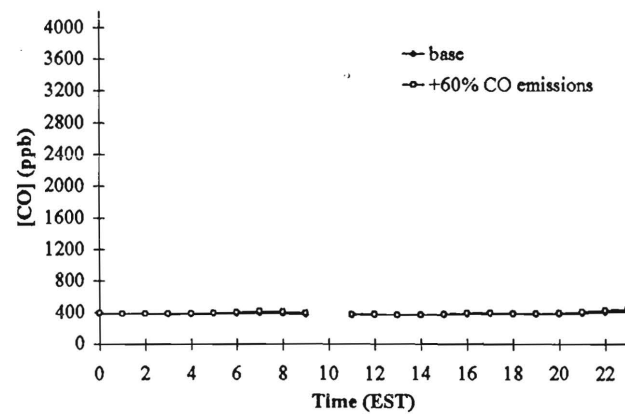
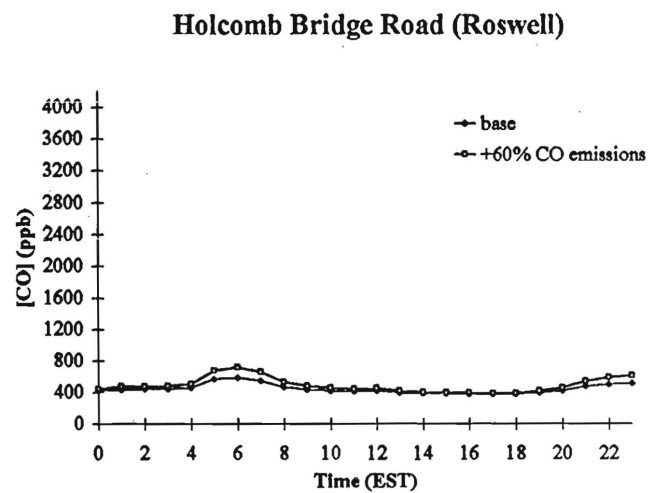
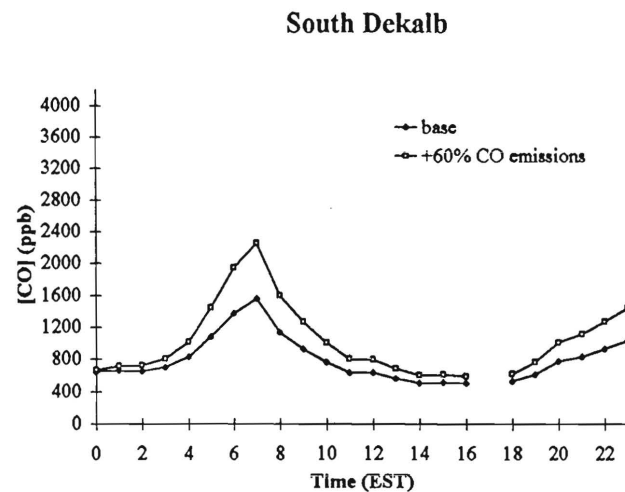
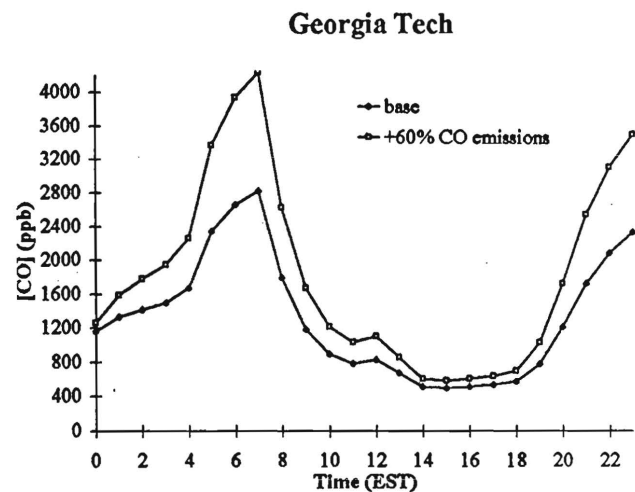


Figure 3. Base (pseudo-observations) and perturbed (+60% CO emissions) UAM predicted CO concentrations in Atlanta, Georgia (August 10, 1992).

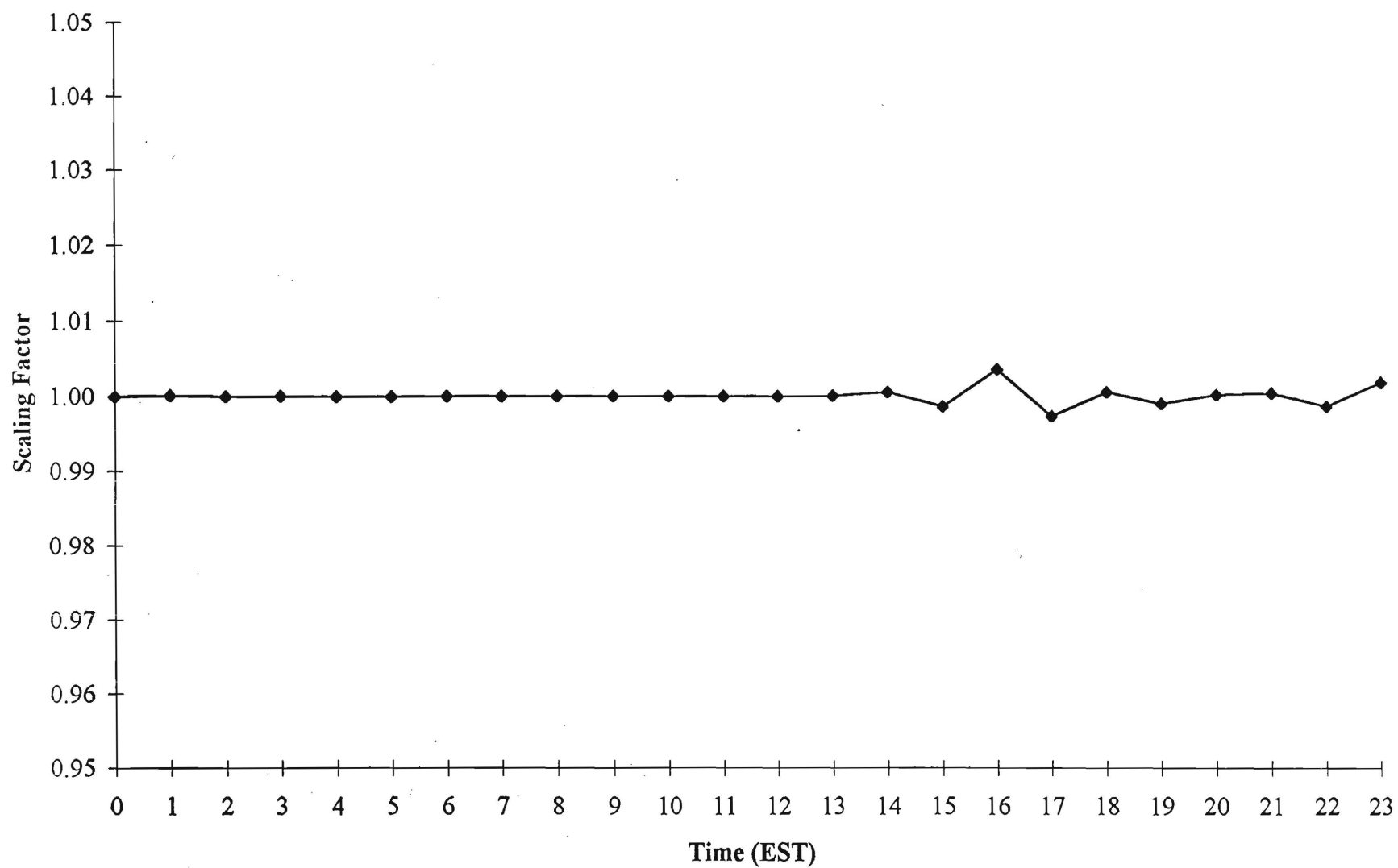


Figure 4. Inverse derived CO emission scaling factors for pseudo-data (60% perturbation).

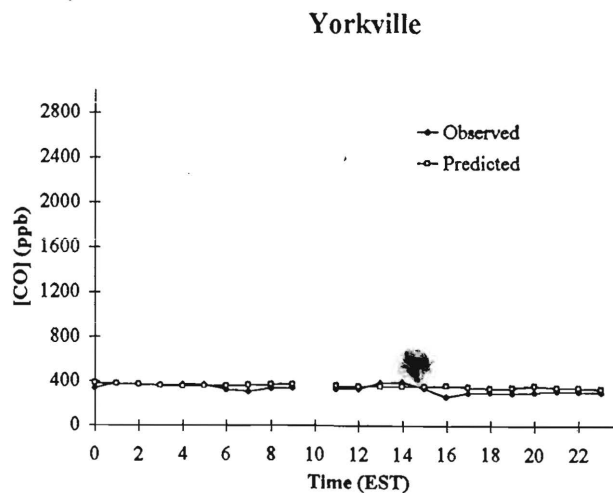
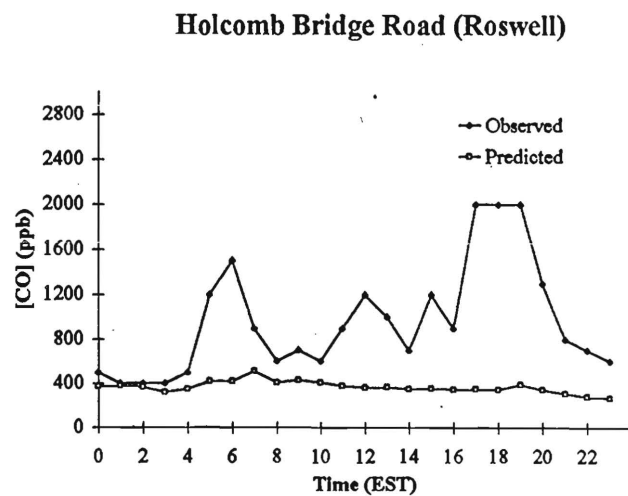
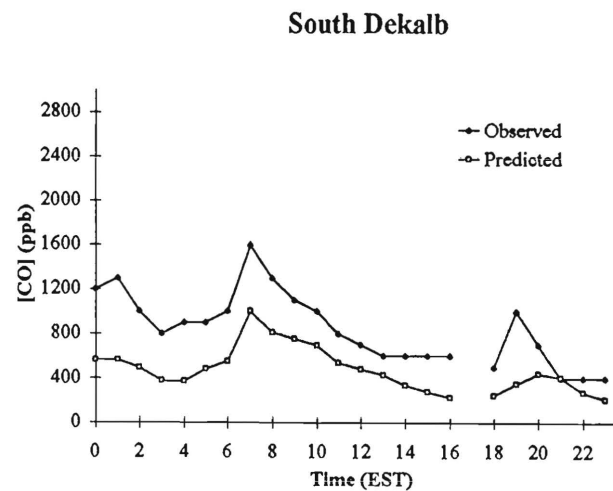
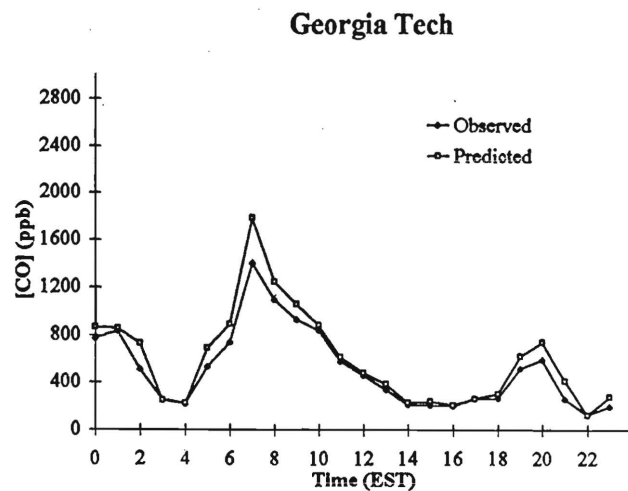


Figure 5. Observed and inverse adjusted - UAM predicted CO concentrations in Atlanta, Georgia (August 10, 1992).

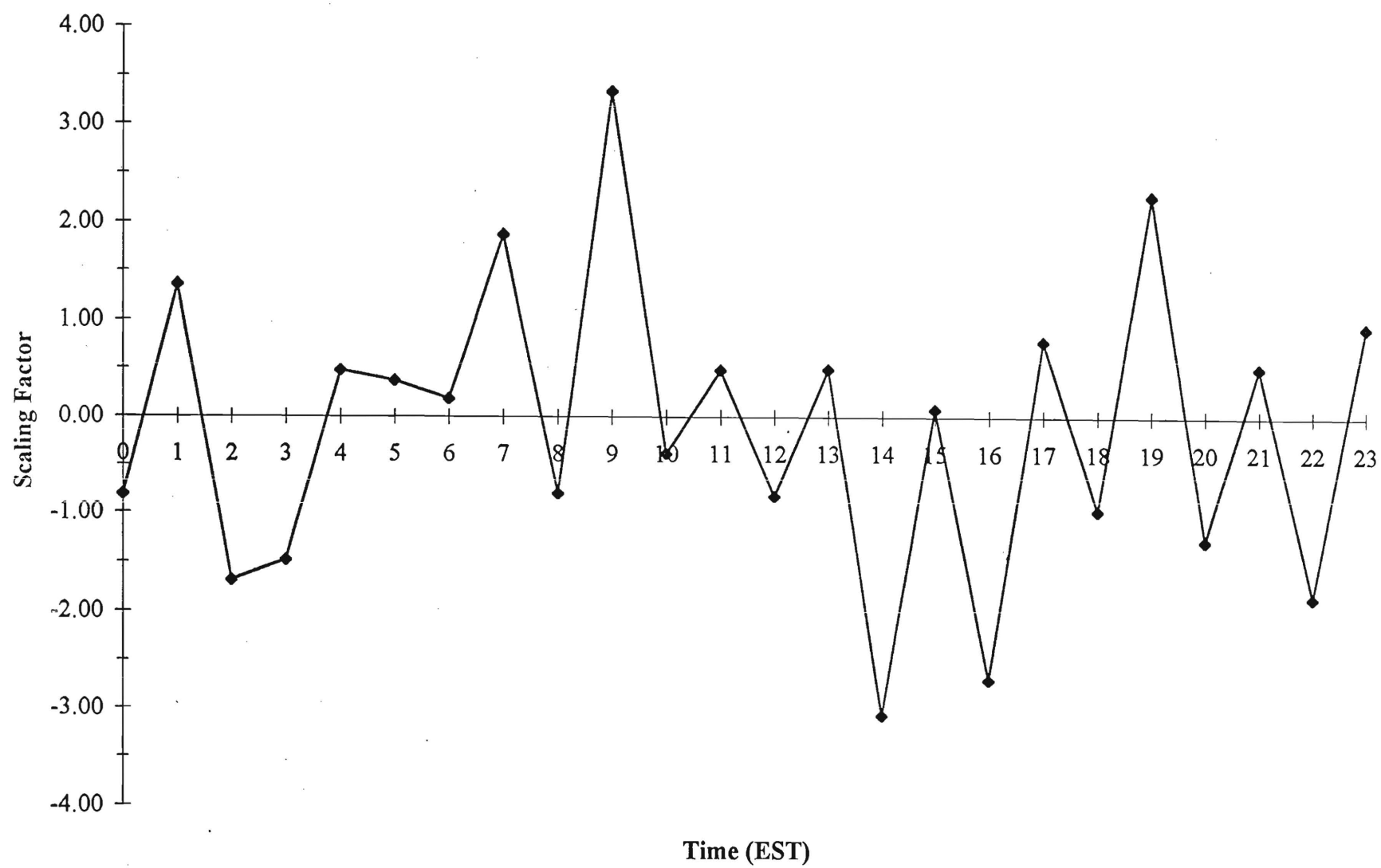
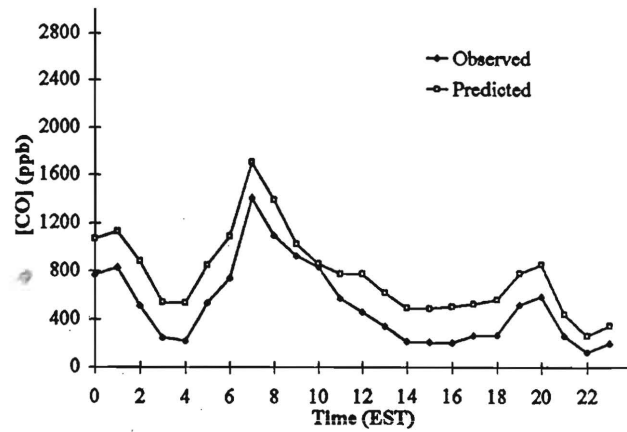
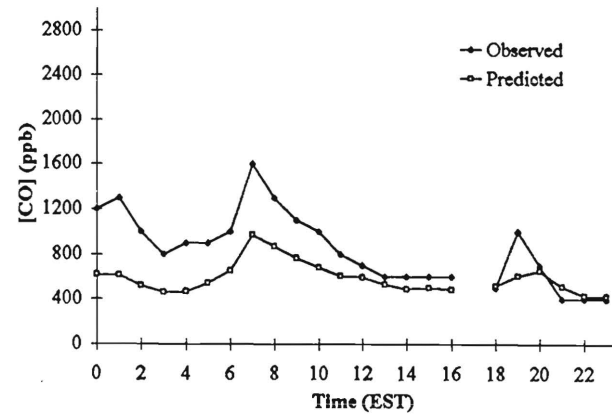


Figure 6. Inverse derived CO emission scaling factors.

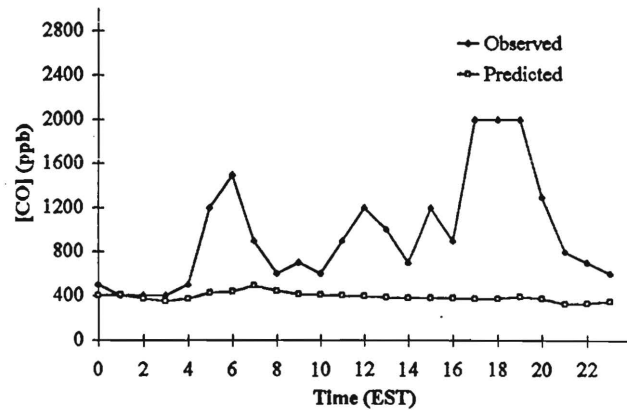
Georgia Tech



South Dekalb



Holcomb Bridge Road (Roswell)



Yorkville

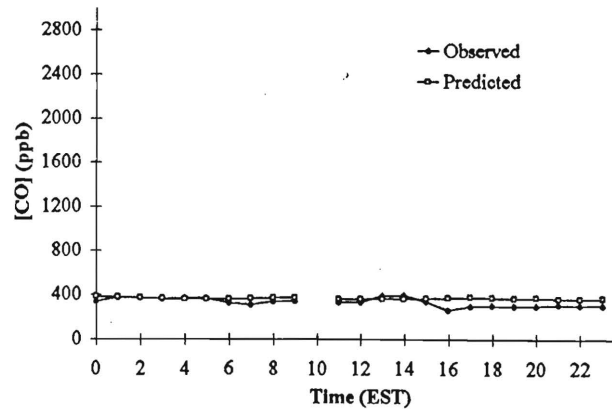


Figure 7. Observed and "tuned" inverse adjusted - UAM predicted CO concentrations in Atlanta, Georgia (August 10, 1992).

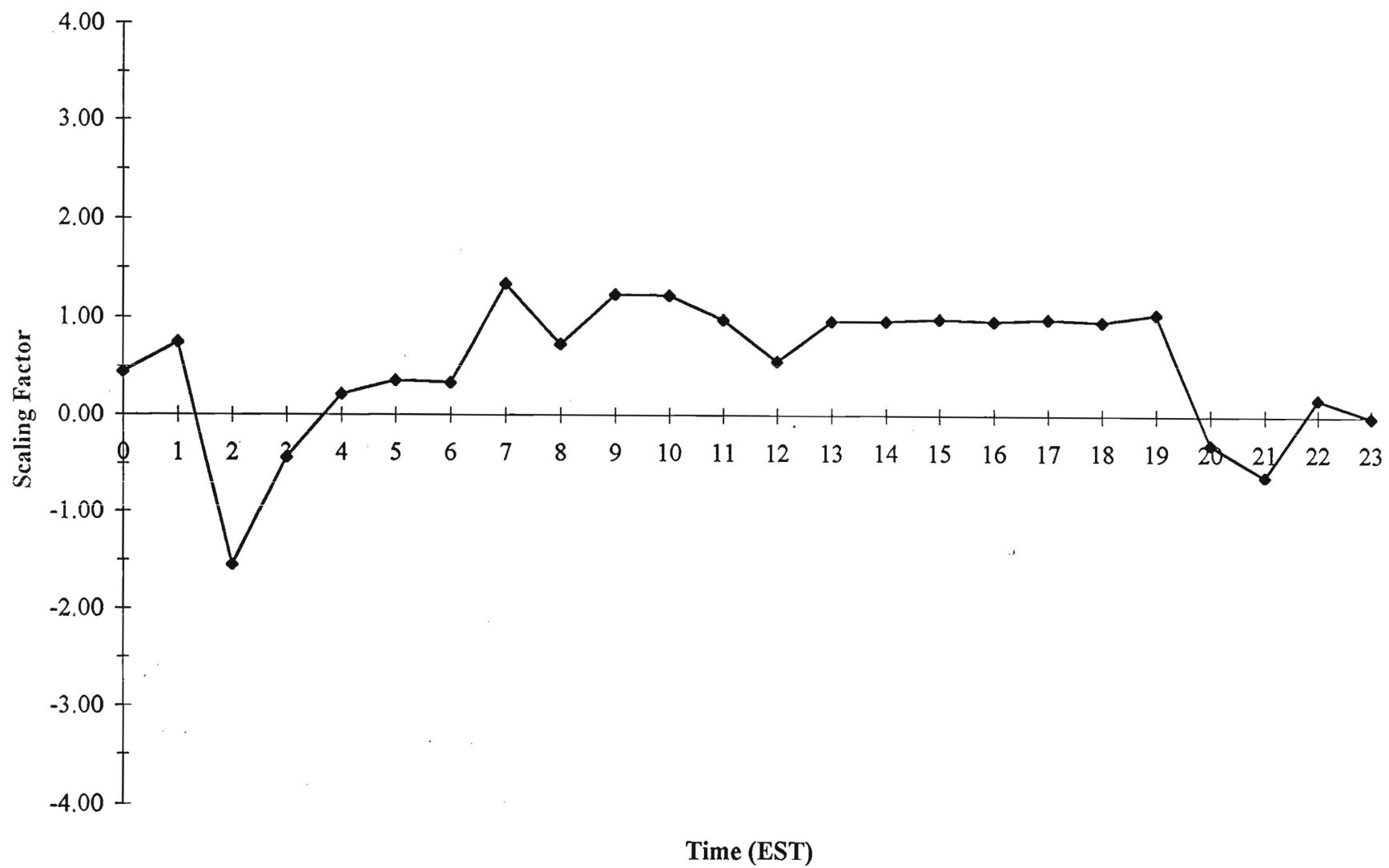


Figure 8. "Tuned" inverse derived CO emission scaling factors.

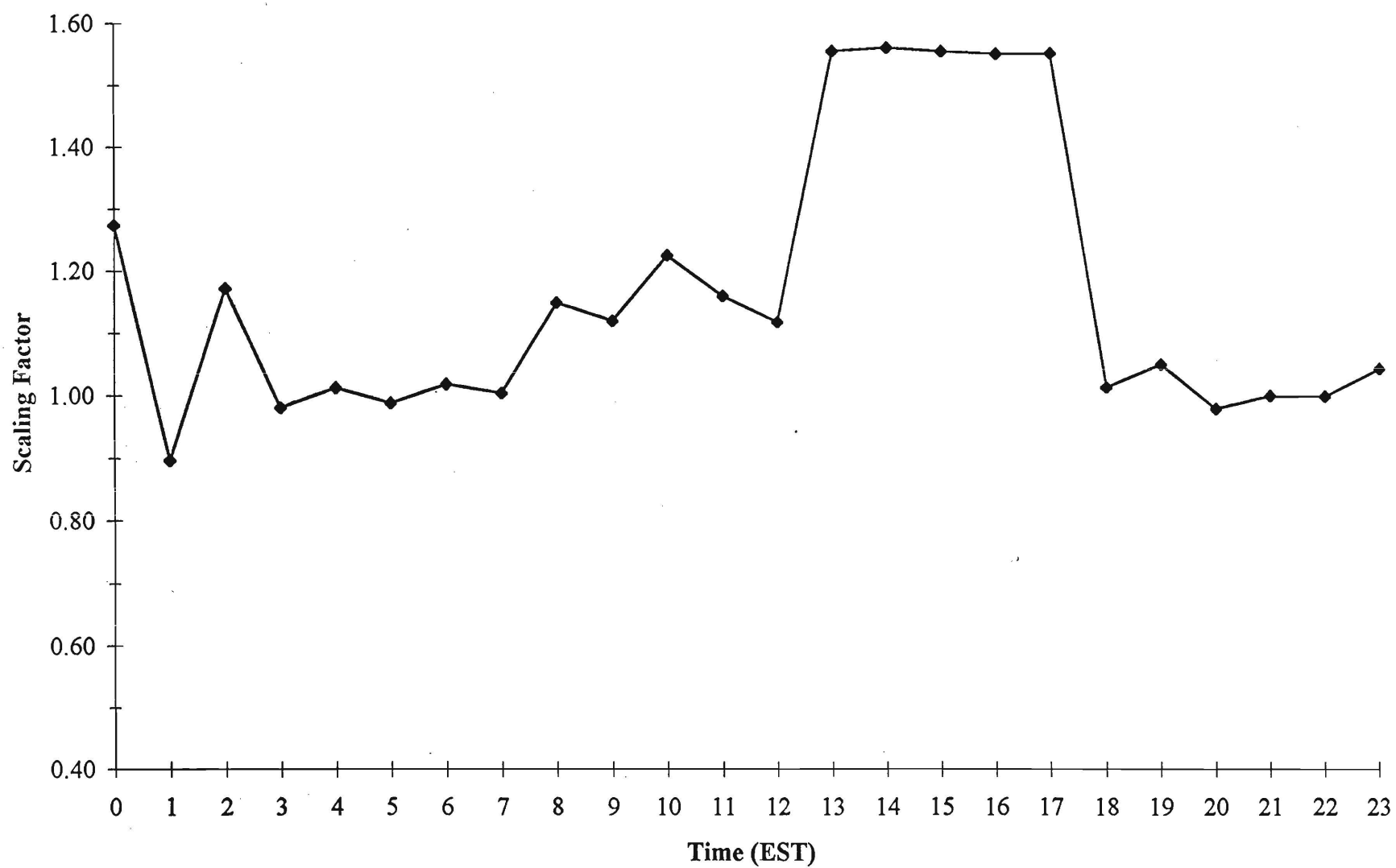


Figure 9. "Tuned" inverse derived CO emission scaling factors for pseudo-data (60% perturbation).

not resolve the emissions at all between hours 13 and 17. This would indicate that the damping effect was too severe. By over-restricting the amount by which the emissions were allowed to change, the emissions could not change sufficiently so that model predictions match the pseudo-observations. This must also be true for the real-data run. Thus, it is difficult to conclude that the encouraging results seen in the "tuned" CO inverse run represent the true CO emissions. Several more runs were made in an attempt to find a compromise between the need to dampen the oscillations and the diametric need to allow sufficient emissions change. However, damping the oscillations proved to be the more difficult to control; they returned as soon as the parameters were relaxed. No satisfactory solution was found.

We suspect that the difficulty in running a CO inverse lies in the spatial distribution of the CO emissions in the modeling inventory. Before we begin an inverse problem, we assume that all the emissions are approximately correctly spatially distributed. Spatial distributions may be determined either directly (e.g. emissions along a roadway) or by surrogate (e.g. CO emissions follow population patterns). Further, while we do not assume that the magnitude of the emissions are correct, we do assume that the relative spatial distribution is correct. For example, if it is known that two times more vehicles travel across the northern arc of I-285 than across the southern arc, then a good first assumption is that the mobile emissions across the northern arc are twice those across the southern arc. Given the spatial distribution, we then solve for the magnitude of the emissions at each hour, thus determining the temporal allocation. In obtaining this solution, we also assume that the meteorology (primarily wind speed, wind direction, and mixing height) is also approximately correct. This plan worked well for the biogenic isoprene emissions. It does not seem to work well for the anthropogenic CO emissions.

In Atlanta, sources of isoprene (all biogenic) are numerous, small, and ubiquitous. There are no highly concentrated isoprene sources. At any isoprene monitor, about the same concentration of isoprene will be measured regardless of the direction of the wind. On the other hand, some sources of CO emissions may indeed be extremely concentrated (e.g. along a roadway). Thus, a CO receptor could measure high concentrations of CO if it is near a source or the wind is blowing from the location of a source. The same receptor could also measure low concentrations if the wind is blowing from another direction. In the model, small errors in the spatial distribution of CO sources or in the speed and direction of the wind could cause large discrepancies between model predictions and observations. Since the biogenics are more homogeneously distributed, this was not a problem during the isoprene inverse.

Last year, we reported (Cardelino et al. 1995) a similar finding using a different approach. For the anthropogenic emissions, we compared observed ratios of HC to NO_x and observed ratios of CO to NO_x to the corresponding ratios derived from emission estimates. The results showed that the ambient ratios of HC/NO_x and CO/NO_x were higher than the emission ratios by 20% and 73% respectively. We also compared UAM predicted concentrations of HC, NO_x, and CO to observed concentrations and found that the ambient concentrations were greater than the corresponding UAM concentrations by 14%, 40% and 58%, respectively. We concluded then that the variability shown by the different comparisons suggested that a possible source of the discrepancies was the spatial distribution of the anthropogenic emissions. Unfortunately with only 4 sites, we do not have enough CO observations to attack the spatial distribution problem with the inverse method.

3. Nitric Oxide (NO)

On August 10, 1992, there were seven (7) nitric oxide (NO) monitors operating in the Atlanta Area. Once again, the Georgia Tech site (TECH) was the only true urban site. The South Dekalb (SDEK), Fort McPherson (FORT), and Tucker (TUCK) sites may be considered as suburban. The rural sites were Yorkville (YORK), Buzzard Flapper (BFCK) and Clover Lake (CLOV). Figure 10 shows the observed concentrations and initial UAM predicted (prior to any adjustments) concentrations.

When observed concentrations are extremely small, the normalized bias (the statistic we have used to measure model performance relative to observations) can become extremely exaggerated:

$$\text{normalized bias} = \{\Sigma(\text{predicted} - \text{observed})/\text{observed}\}/N$$

where N is the number of prediction-observation pairs.

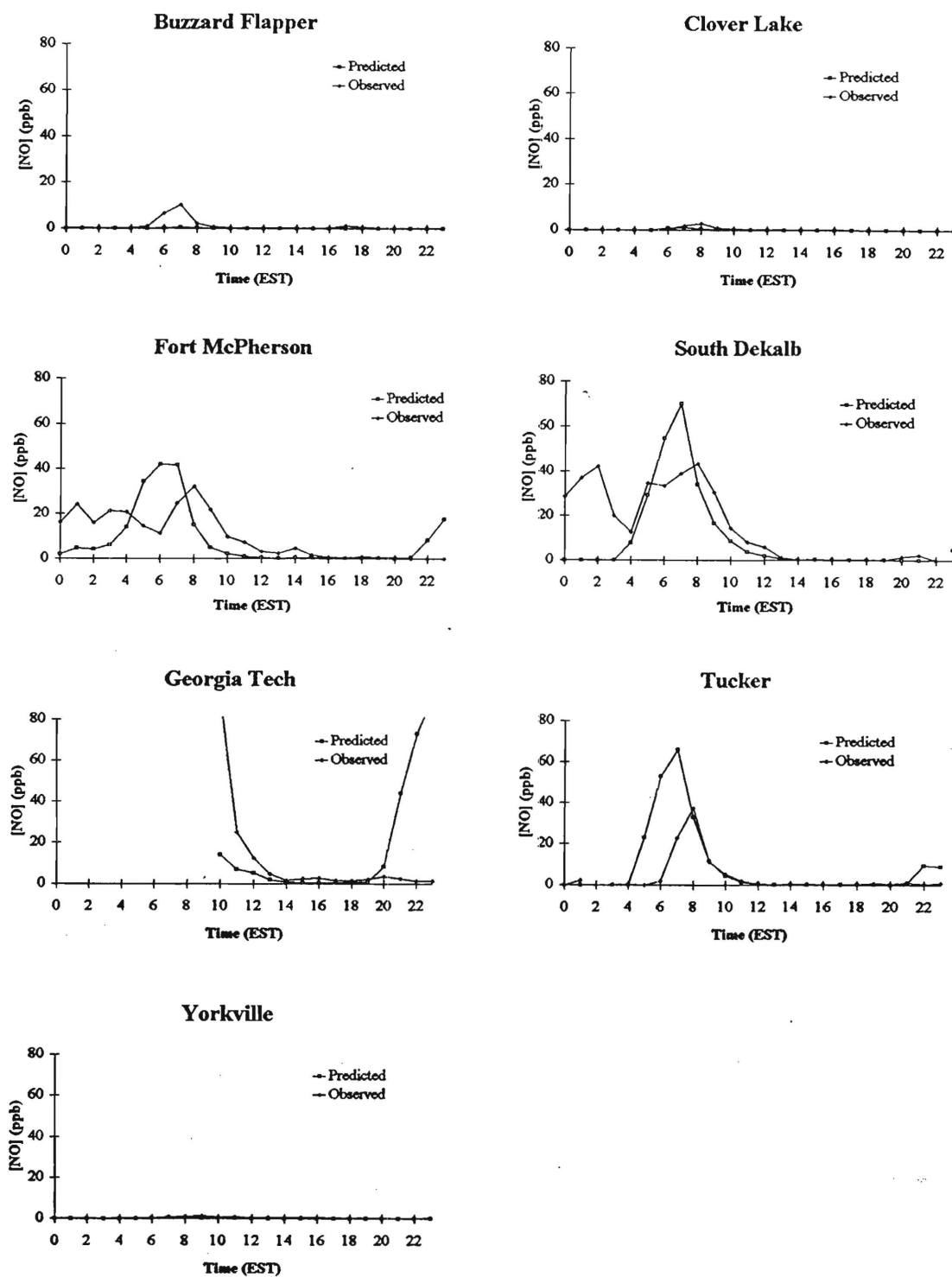


Figure 10. Observed and initial UAM predicted NO concentrations in Atlanta, Georgia (August 10, 1992).

In these cases, it is more appropriate to use the bias to assess model performance:

$$\text{bias} = \{\sum(\text{predicted} - \text{observed})\}/N$$

The latter is actually what the inverse method is attempting to minimize. Overall, before any adjustments, the UAM slightly overpredicts NO concentrations by 0.18 ppb. Performance is the worst at the TECH site; at this site, on average the model overpredicts NO by 6.91 ppb. There are also differences at the suburban sites. The model underpredicts by 1.42 ppb at the FORT site and by 5.24 ppb at the SDEK site. It overpredicts by 5.20 ppb at the TUCK location. The model predicts NO relatively well for the rural sites. The biases at BFCK, CLOV, and YORK are +1.04 ppb, -0.20 ppb, and -0.34 ppb respectively.

Since the model is already performing relatively well for most hours as evidenced by the small overall bias, it will be difficult to improve the simulation using the inverse method. In particular, the afternoon hours will be extremely difficult since concentrations of NO do not seem to be affected by emissions. Further, NO emissions also share some of the same spatial attributes (and thus some of the same problems) as CO emissions. Nevertheless, we did attempt to run an inverse for NO.

A. NO Inverse with Pseudo-data

All of the area, nonroad mobile, and mobile NO emissions sources were merged into one gridded and temporally allocated source. Although point sources are a significant source of NO in the Atlanta area, it was assumed that their emissions were well known in the 1992 inventory. This is a valid assumption since day specific data was used for 90% of the total point source NO_x emissions (Chang et al. 1996). For the inverse, it must also be assumed that the UAM adequately simulates point source plumes. (This is a widely noted deficiency in version IV of the UAM, and thus another obstacle for the NO inverse. Plumes are more aptly treated in version V of the model with a plume-in-grid mechanism.) The single low-level source was initially set to 150% of the baseline. As with the CO run, the filter was then used to determine the base emissions using only the differences between the predicted NO concentrations (with initial emissions at 150% of the baseline) and the observed concentrations (pseudo-data - NO concentrations extracted from a baseline run). Figure 11 shows the pseudo-data observations and the initially perturbed concentrations at each of the 7 sites. Figure 12 shows the inverse derived factors necessary to force the model to match the pseudo-data. Using pseudo-data, we expected to get a near perfect solution (recall that a value of 1.00 implies a perfect convergence), yet the solution was less than perfect. This can indicate that the NO signal is not strong enough to overcome the difficulties associated with running an inverse for NO. From this test with an induced 50% error in the emissions, we decided to cautiously proceed with a real-data NO inverse run. Any results obtained must be viewed in the context of the uncertainties described above.

B. NO Inverse with Observations from the 1992 Atlanta Intensive

NO observations from the 1992 Atlanta Intensive (see Figure 10) were used to determine hourly factors needed to adjust the August 10, 1992 (day specific) NO emission inventory to minimize the differences between model predictions and observations. This run was terminated after hour 10 when it was noticed that an unusually large number of iterations were required before the solution would converge. Typically, the method will converge on a solution in less than 10 iterations. For hour 3, the method required 268 iterations. Nevertheless, this abbreviated run does provide some insight. Figure 13, when compared to figure 10 shows that the inverse was functioning properly - model predictions at each station tended to follow the observed profiles more closely than in the initial unadjusted run. For hours 0-10, the bias at most sites decreased. For BFCK, CLOV, FORT, SDEK, TECH, TUCK, and YORK the inverse adjusted bias statistics (and original unadjusted bias statistics) are -1.90 (-1.91), -0.30 (-.32), +1.61 (-3.75), -13.63 (-10.32), -53.69 (-76.03), +5.81 (+10.95), -0.32 (-0.33) respectively. Even at the South Dekalb site where the statistics indicate that the simulation was made worse by the adjustment, visual inspection suggests that the inverse simulation better represents the observations.

The inverse derived emission factors are shown in figure 14. Although the run was terminated prematurely, results from hours 7-10 indicate that the solution may again oscillate from positive to negative emissions similar to the CO inverse. When the run was terminated during hour 11, emissions were moving back towards a negative

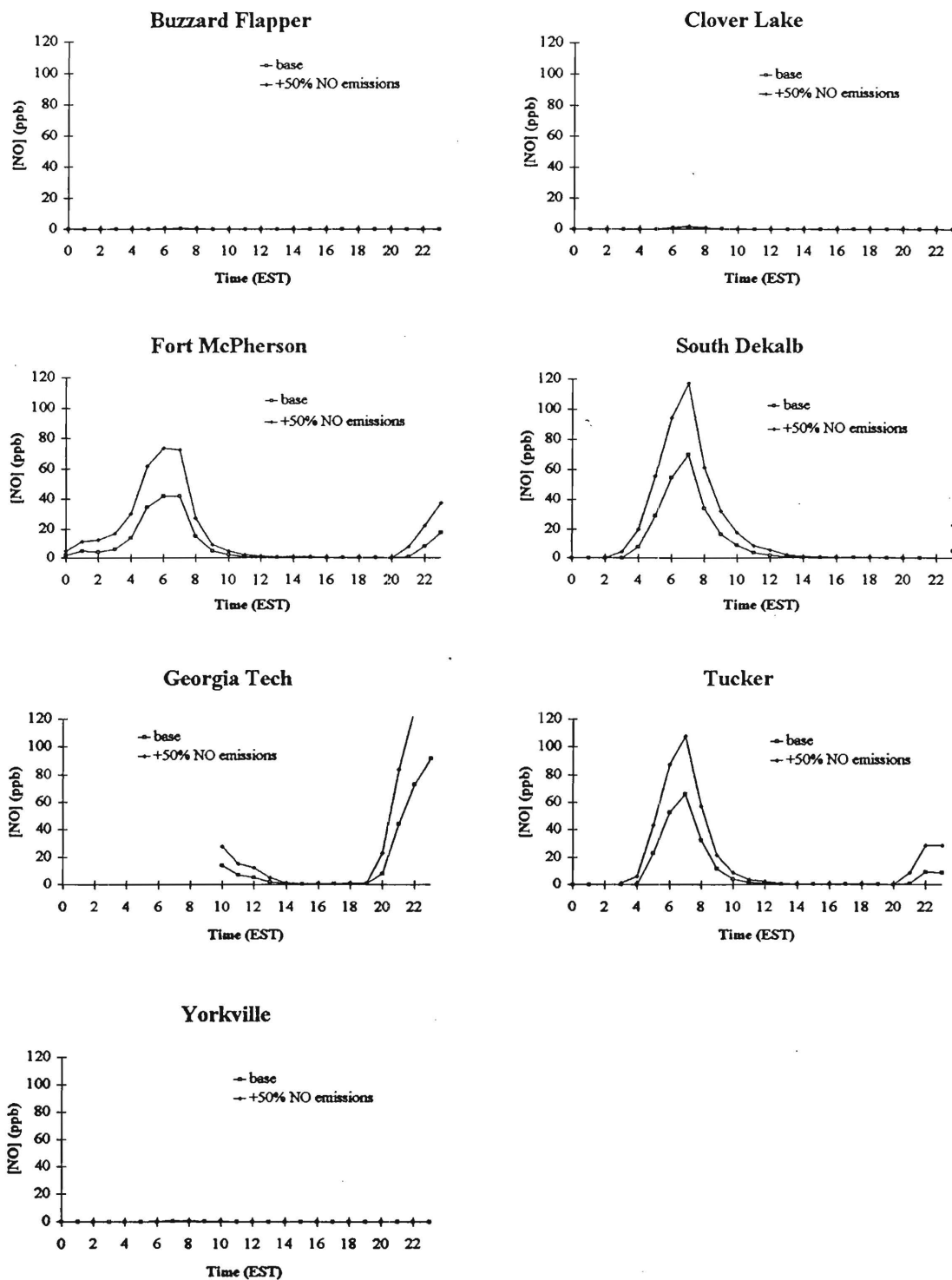


Figure 11. Base (pseudo-observations) and perturbed (+50% NO emissions) UAM predicted NO concentrations in Atlanta, Georgia (August 10, 1992).

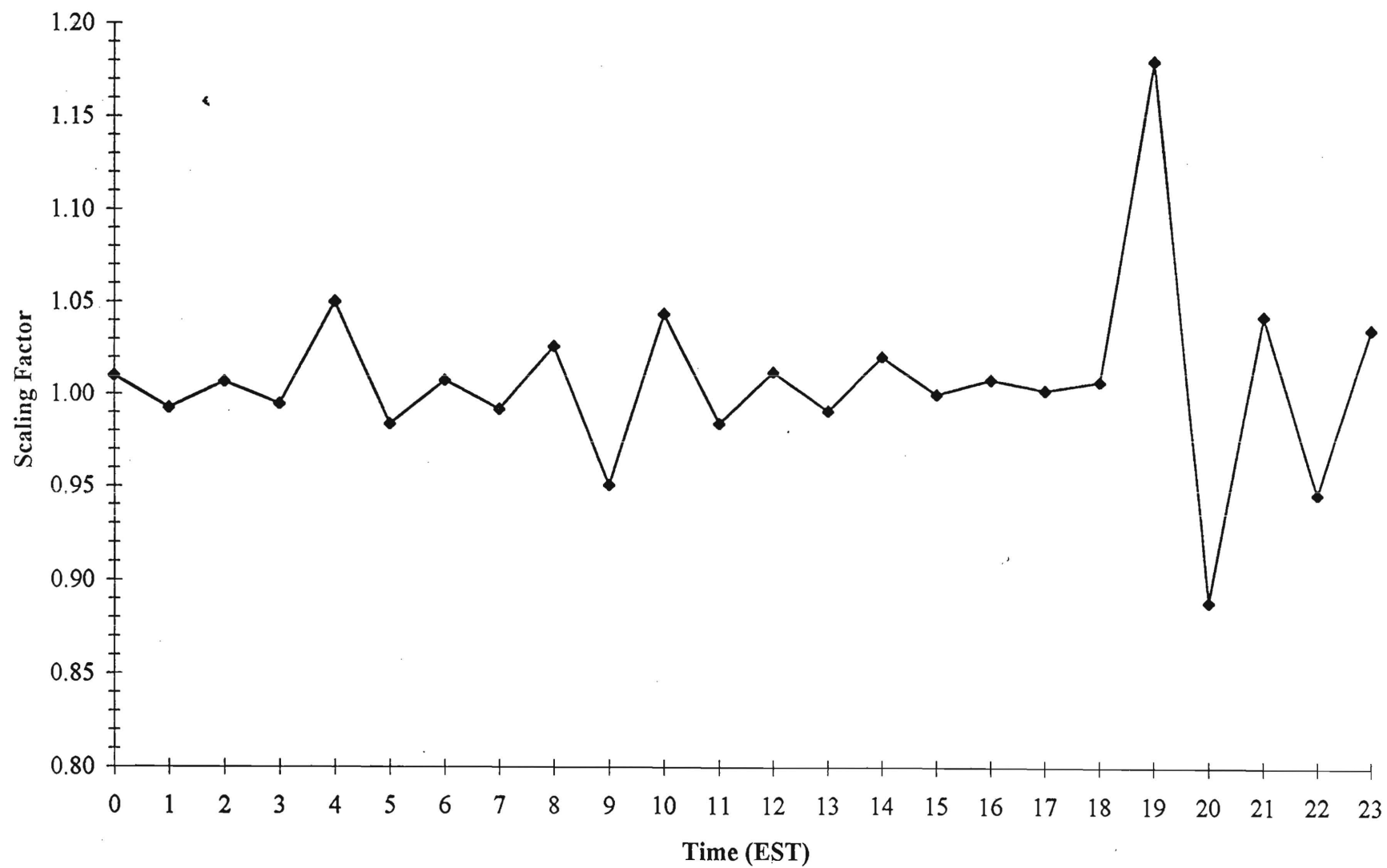


Figure 12. Inverse derived NO emission scaling factors for pseudo-data (50% perturbation).

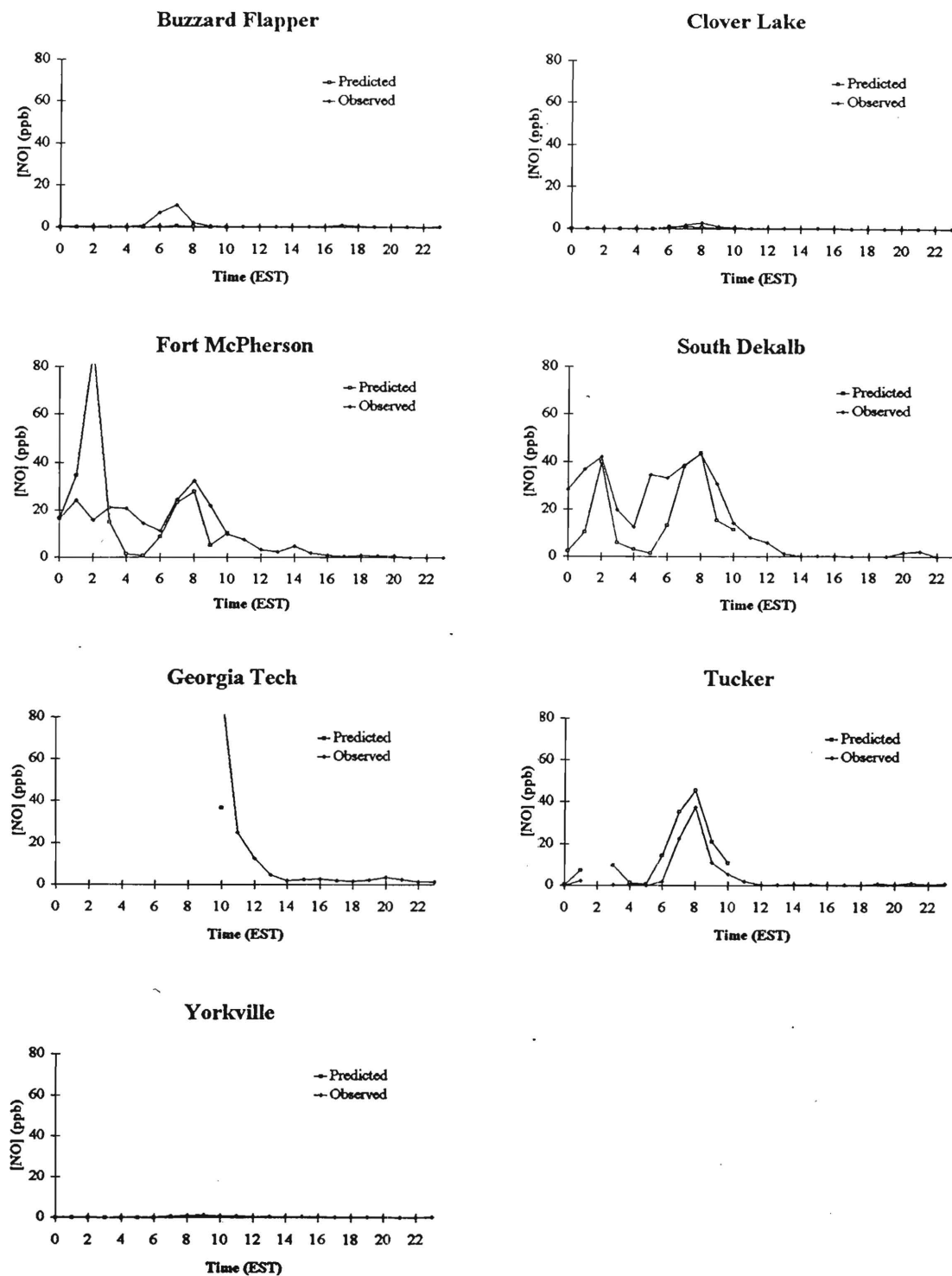


Figure 13. Observed and inverse adjusted - UAM predicted NO concentrations in Atlanta, Georgia (August 10, 1992).

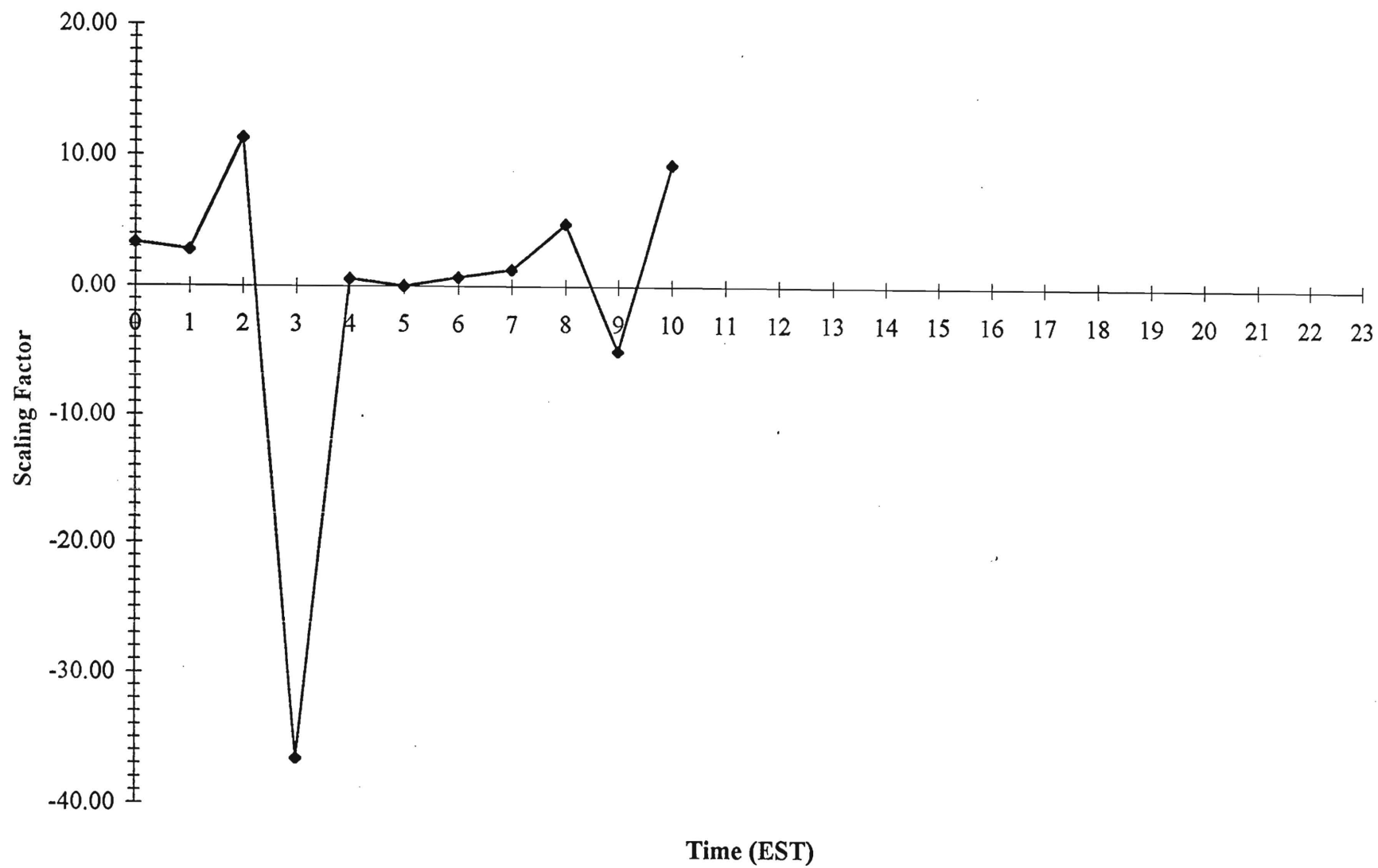


Figure 14. Inverse derived NO emission scaling factors.

solution (not shown) following the positive adjustment during hour 10. This should not be surprising given that the low-level NO emissions are dominated by the same sources (i.e. mobile) as the CO emissions. We would expect that the NO inverse would have many of the same problems as the CO inverse. Given this sign along with the other difficulties described previously, no further NO inverse runs were completed.

4. Volatile Organic Compounds (VOCs)

Relative to the inverse problem to estimate the emissions of anthropogenic hydrocarbons, the isoprene, carbon monoxide, and nitric oxide inverse problems should have been easy to solve. In each of the previous cases, the chemical specie studied was measured explicitly and modeled explicitly. Within Atlanta, the only significant source of isoprene is biogenic emission; the dominant sink is reaction with the hydroxy radical (OH). Unlike the other hydrocarbons, there is no in situ production due to the oxidation and reaction of other hydrocarbons. Thus, isoprene works well in the inverse method because there is very little interference from other mechanisms. Likewise, we did not expect to have much difficulty with carbon monoxide. On the time scale of 1 day, CO is virtually unreactive. Further, since the emission of CO is dominated by only one source (mobile), using the inverse to solve for the CO emissions should have been relatively straight forward. We did not anticipate the difficulties associated with the spatial allocation of the sources. We did anticipate that NO would provide additional problems. Although the majority of NO_x emitted is in the form of NO, during the day it is quickly oxidized so that the majority of ambient NO_x is in the form of NO₂. This property makes it difficult to measure the change in ambient NO concentrations due to a change in NO emissions. Still, since NO_x chemistry is treated explicitly in the UAM, this should have been a minor obstacle.

Because anthropogenic hydrocarbons are neither explicitly measured nor explicitly modeled, substantial noise is introduced into the inverse method. Literally hundreds of hydrocarbons are present in the ambient air. Only 56 of these species were measured during the 1992 Atlanta Intensive. It is not clear if this small sample is representative of the total volatile organic compound loading that existed in Atlanta. Further, to facilitate comparison with model predictions, these observations must first be mapped into the Carbon Bond IV (CB-IV) chemical mechanism. This will further degrade the quality of the measurements by subjecting them to the vagaries of the chemical model. From the emissions side, estimates of total VOC must also undergo the CB-IV transformation. While none of these issues would have prevented us from conducting an inverse analysis of the anthropogenic VOCs, these together with the difficulties we discovered in trying to solve for the CO and NO emissions have led us to believe that it would be fruitless to attempt a VOC inverse. We believe that failure to find a satisfactory inverse solution for the CO and NO inventories is a sufficient reason to conclude that a suitable VOC inverse solution also does not exist.

5. Future Plans

Over the next several months, we will test other approaches to determine if we can minimize the problems associated with the spatial allocation of the anthropogenic emissions. Specifically, we will experiment with longer averaging times (e.g. 3 hours instead of 1 hour), and a time inclusive approach rather than a time iterative approach. This work will be submitted to a peer review journal and we will continue to keep the DNR informed of any progress.

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- Chang, W.L., C. Cardelino, M.E. Chang; The Use of Survey Data to Investigate Ozone Sensitivity to Point Sources; in press *Atmospheric Environment*; 1996.
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III. Appendix

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