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Evaluation of Ultraviolet Radiation, Ozone and Aerosol Interactions in the Troposphere using Automatic Differentiation

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1. Project Overview

Interactions between ozone, aerosols, and ultraviolet radiation are investigated using a detailed radiation model combined with a three-dimensional atmospheric chemistry model. These models are joined with Automatic-Differentiation software to enable desired sensitivities to be calculated on-line with the radiation/chemistry computations. The combined methodology was used to investigate the following:

(a) UV-B radiation at the earth's surface as a function of changes in ozone and aerosols in the stratosphere and troposphere;
(b) effects of changes in solar actinic flux on the photochemical oxidant cycle of the troposphere.

The project team consisted of Gregory R. Carmichael, Professor, Department of Chemical & Biochemical Engineering and Co-Director of the Center for Global & Regional Environmental Research; Florian Potra, Professor of Mathematics and Computer Science; and a group of graduate students from Chemical & Biochemical Engineering, Applied Mathematics and Computer Science. The work focused on three areas: investigations of the couplings between tropospheric chemistry and radiative transfer using the STEM-II model; implementation and use of automatic differentiation; and improvements in the numerical integration of the models.

A major goal of this research was to quantify the interactions between UVR, ozone and aerosols. One method of quantification was to calculate sensitivity coefficients. A novel aspect of this work was the use of Automatic Differentiation software to calculate the sensitivities. We demonstrated the use of ADIFOR for the first time in a dimensional framework. Automatic Differentiation was used to calculate such quantities as: sensitivities of UV-B fluxes to changes in ozone and aerosols in the stratosphere and the troposphere; changes in ozone production/destruction rates to changes in UV-B flux; aerosol properties including loading, scattering properties (including relative humidity effects), and composition (mineral dust, soot, and sulfate aerosol, etc. The combined radiation/chemistry model offers an important test of the utility of Automatic Differentiation as a tool in atmospheric modeling.

The combined sensitivity analysis with detailed radiation and atmospheric chemistry modeling is computationally very complex. In recognition of this fact, and to take advantage of the strengths of our interdisciplinary research group, we also addressed several computational issues. The primary focus is on the computational considerations of ADIFOR. However the accuracy of the ADIFOR sensitivities depends on the numerical methods used in the STEM-II and the radiation code. Therefore we investigated ways to improve the performance (speed and accuracy) of the key algorithms in the models. Specific activities and results are discussed below.

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2. Project Results

2.1 Sensitivity Calculations using ADIFOR

A variety of techniques are available for calculating sensitivities ranging from brute force, to Green's function analysis, and to several variations of the direct decoupled methods. Most sensitivity studies with comprehensive atmospheric chemistry/transport models have been done using the so-called "brute force" method, i.e., a number of input parameters are selected to be varied and the simulation results are then compared. This method becomes less viable as the model becomes more comprehensive. Some work has been done using sensitivity analysis embedded in the calculation itself (i.e., an on-line mode). For example we have used sensitivity methods by Rabitz to study a variety of atmospheric transport and chemistry issues. However these studies have not included aerosols and have not been fully exploited.

A recently developed technique for sensitivity study is automatic differentiation. Automatic differentiation is implemented by precompilers that analyze the code written for evaluating a function of several variables. These precompilers automatically add instructions needed to compute the required derivatives by properly handling quantities that are common to the function and its derivatives and by efficient use of available derivatives in a library. The resulting expanded code is then compiled with a standard compiler into an object code that can simultaneously evaluate derivatives and function values. This approach is, by far, superior to finite difference approximation of the derivatives because the numerical values of the computed derivatives are much more accurate and the computational effort is significantly lower.

A new implementation developed at Argonne National Laboratory and Rice University over the last couple of years is the package ADIFOR (Automatic Differentiation in FORTRAN). It adopts a hybrid approach to computing derivatives that is generally based on the forward mode, but uses the reverse mode to compute the derivatives of assignment statements containing complicated expressions. The forward mode acts similarly to the usual application of the chain rule in calculus. At each computational stage derivatives of the intermediate variables with respect to input variables are computed and are propagated forward through the computational stages. ADIFOR requires the user to supply the FORTRAN source code for the function value and for all lower level subroutines as well as a list of the independent and dependent variables in the form of parameter lists or common blocks. ADIFOR then determines which other variables throughout the programs are to be differentiated, and augments the original code with derivative statements. The augmented code is then optimized by eliminating unnecessary operations and temporary variables. The FORTRAN code generated by ADIFOR requires no run-time support and therefore can be ported between different computing environments.

In this research we used ADIFOR for the computation of the sensitivities. It is easy to use and supports almost all of FORTRAN, including nested subroutines, common blocks and equivalencies so that very few modifications are needed in the original FORTRAN code. Moreover, the derivative code offers additional possibilities for vectorization and parallelization.

In our first study using AD we calculated the sensitivity of O_3, OH, HO_2, H_2O_2 to all initial conditions and all chemical reaction rate constants for the IPCC gas phase chemical mechanism test scenarios (6 cases). Six different scenarios were simulated which represent atmospheric chemistry conditions ranging from marine, to free-troposphere, to urban plumes. Simulations were performed for 5 days, with each day containing a diurnal cycle. The details are presented in a paper in *Atmospheric Environment* (Sandu et al., 1996b). The calculated ozone concentrations for each case are presented.

ADIFOR was used to calculate the sensitivity of ozone (and eight other important species) to the initial conditions of 81 species and all 178 reaction rate constants. The results for case-6 at the end of 5-days are shown for illustration in **Table 1**. Shown are the normalized local sensitivities; i.e.,

$$s^{*}_{i,j} = \frac{\alpha_j}{c_i} \cdot s_{i,j} = \frac{\partial \ln(c_i)}{\partial \ln(\alpha_j)} = \frac{\frac{\Delta c_i}{c_i}}{\frac{\Delta \alpha_j}{\alpha_j}}$$

The individual sensitivities are further presented in terms of the positive and negative influence (- meaning that the concentrations decrease with an increase in that parameter). In the urban plume case we find that the largest (+) initial condition sensitivities come from NO_X, ethene, and O_3; while the largest (-) sensitivities come from aldehydes, higher alkenes, and aromatics. The largest (+) rate constant sensitivities come from the reactions: NO_2 + hv; MCO_3 + NO; ALD2+ hv; and ETHE + OH; while the largest (-) sensitivities come from the reactions: $MCO_3 + NO_2$; $NO + O_3$; $O_3 + HO_2$; and ALD2 + OH.

The use of AD in coupled chemistry/radiation calculations is discussed later in this report (Section 2.3).

2.2 Improved Chemical Integrators

Although AD calculates derivatives which are exact up to the machine precision, the accuracy of the derivatives of the time-dependent concentrations depends on the accuracy of the numerical integration scheme. In fact, the errors of the numerical scheme will dominate those induced by the AD process. As one major focus area in this project, we developed techniques that can provide more accurate solution with lower computational efforts for the integration of the chemistry equations when used with forward mode AD.

Table 1. Calculated normalized sensitivities of ozone and trace species with respect to initial values for the 6 IPCC scenarios. Shown are results after 5 days.

$\hat{L}(...)$	Marine	Land	Bio	Free	Plume-1	Plume-2
O_3	8.18E - 01	5.41E - 01	6.40E - 01	9.25E - 01	3.93E - 01	1.52E - 01
CH_4	5.41E - 02	1.16E - 01	8.13E - 02	9.27E - 03	9.89E - 02	8.70E - 03
CO	-1.04E - 02	3.41E - 02	4.05E - 03	-5.04E - 03	0.31E + 00	4.48E - 02
HNO_3	4.82E - 02	2.84E - 02	3.00E - 02	5.74E - 03	9.20E - 04	1.55E - 03
NO_2	1.14E - 02	7.08E - 02	1.10E - 01	1.52E - 02	-7.84E - 02	3.23E - 01
NO	1.14E - 02	6.97E - 02	1.09E - 01	1.46E - 02	-1.17E - 01	3.05E - 01
H_2O_2	-1.12E - 02	-7.63E - 03	-5.89E - 03	-7.32E - 03	9.03E - 02	-5.04E - 03
ISOP	–	–	-7.50E - 02	–	–	-1.36E - 02
ETHE	–	–	–	–	–	1.64E - 01
ALD_2	–	–	–	–	–	-1.39E - 01
ALKE	–	–	–	–	–	-9.34E - 02
AROM	–	–	–	–	–	-6.36E - 02
ALKA	–	–	–	–	–	-2.51E - 02
C_3H_8	–	–	–	–	–	-1.54E - 02

It is well known that the chemistry rate equations comprise a system of stiff ordinary differential equations (ODE). This fact precludes the use of traditional explicit integration schemes. Two families of methods are usually employed to solve the chemical rate equations in atmospheric chemistry models:

1. Dedicated methods. Obtained by carefully exploiting the particular properties of the model, all these methods try to avoid the numerical cost of quasi-Newton iterations which appear in classical implicit methods. Some are based on the production-destruction form (QSSA), Twostep, others on special partitionings (Sillman), EBI method, hybrid method of Gong and Cho, and the projection/forward differencing method of Elliot et al..

2. General implicit methods (e.g., Jacobson et al.).

We studied in detail both families of methods. From the dedicated methods, we have studied the QSSA method which is widely used in atmospheric chemistry studies (Jay et al., 1996). The mathematical insight allowed us to propose two new (and better) methods in this class, namely the Extrapolated QSSA Method and the Symmetric QSSA Method. Both perform better (factor of 3-5 speed-up) as compared to the classical QSSA scheme.

Our contributions to the use of classical implicit methods in atmospheric chemistry models are presented in (Sandu et al., 1996a, 1996b, 1996d). We have shown that implicit methods can be efficient (and the method of choice) if the linear algebra is properly treated. The advantages of using implicit methods are clear: unconditional stability (for problems involving inter-phase mass transfer explicit methods may completely fail), more robustness, higher accuracy, and mass conservative. For efficiently treating sparsity we developed our own linear algebra routines, based on an off-line analysis of the sparsity structure, a diagonal Markowitz reordering of the species, a Doolittle decomposition and a loop free forward-backward substitution (all implemented within the KPP preprocessor http://www.cgrer.uiowa.edu/people/vdamian). In Sandu et al. (1996d) we proposed two new numerical methods of Runge-Kutta-Rosenbrock type, viz. ROS3 (are L-stable, third order formula with an embedded strongly A-stable formula of order 2 for step-size control; 3 stages but only 2 function evaluations) and RODAS3 (a stiffly accurate order 3 method, with a stiffly accurate order 2 embedded method for step-size control; 4 stages but only 3 function evaluations). In Sandu et al. (1996c, 1996d) we performed a comprehensive benchmark of the best available ODE solvers for atmospheric chemistry. Results were obtained for a large number of test problems and chemical conditions. **Figure 1** (left) presents the work-precision diagram for an urban scenario, modelled with CBM-IV chemical mechanism. One conclusion of the benchmark study is that, under the tested conditions, Rosenbrock methods (and especially RODAS3) are very well-suited for atmospheric chemistry applications.

The development of chemical schemes for heterogeneous chemistry modeling puts new demands on the quality of numerical schemes. For these models the traditional integrators (e.g. QSSA) may prove useless. One example is the gas-and-liquid system from (Sandu et al., 1996c) on which all explicit integrators fail (because of stability problems). On the other hand, as seen in **Figure 1** (right), all (semi) implicit integrators solve this problem very efficiently. The new multiple phase chemical models raise new issues. For example, modeling the gas-aerosol equilibria by algebraic relations transforms the ODE (ordinary differential equation) system into a DAE (differential-algebraic equation) system and special techniques to numerically simulate the time evolution of such a constrained system has to be developed for this application. In particular we need to design special, highly efficient DAE solvers for use in heterogeneous chemistry modeling.

2.3 Ozone-UVB Interactions

Several studies on the interactions between ultraviolet radiation (UVR) and atmospheric chemistry have been performed. One study investigated the effects of changes in atmospheric ozone on surface UVB. In this study a one-dimensional radiative code with high wavelength resolution is used to analyze changes in radiative transfer due to negative trends in stratospheric ozone. This analysis included the calculation of changes in ground level radiative flux as a function of changes in stratospheric and tropospheric ozone levels.

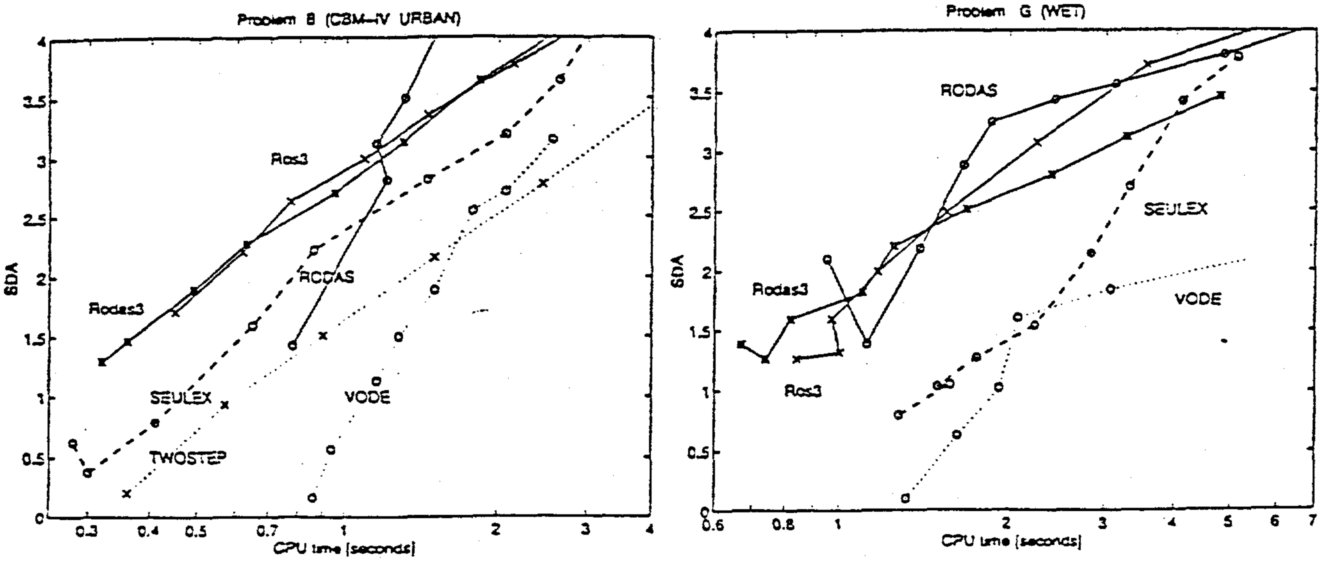


Figure 1: Work-precision diagram for two different tests: an urban scenario with the gas-phase CBM-IV chemical mechanism (left) and an urban scenario with a gas (CBM-IV)-and-liquid mechanism (right). Results for different stiff ode integrators are presented. No explicit integrator is able to efficiently solve the gas-and-liquid problem (instability). The (semi) implicit schemes use sparse implementations of the linear algebra.}

We looked at TOMS trends at mid latitudes over the midwest and found a 5.5 percent annual averaged decline in stratospheric ozone per decade. Simulations of radiative transfer using these trends showed a significant increase in ground level UVR flux as the result of stratospheric ozone reduction. It was shown that this increase in radiative flux at the surface was not uniform, with larger increases occurring in the shorter wavelengths. This enhanced response in the shorter wavelengths results in a large increase in the DNA dose.

An analysis of the effects of tropospheric ozone on radiative transfer for the eastern United States showed that tropospheric ozone is a disproportional filter of solar radiation. Here we used a regional scale model to simulate tropospheric ozone in the eastern United States. These tropospheric ozone profiles were then used in the radiative transfer model. It was found that tropospheric ozone increases in urban areas reduces the flux of UVB at the surface and thus can offset increases in UVB due stratospheric ozone loss. This offset results in a masking of potential ground level UV-B increases.

We also investigated various interactions between radiative transfer and the regional-scale photochemical oxidant cycle. Here we combined the radiative transfer model with the STEM-II model. The radiative transfer model was used to calculate photolysis rates, under a variety of conditions reflecting changes in stratospheric ozone, used in the STEM-II model.

Results using the radiation code indicate that tropospheric photolytic dissociation rates would not change linearly with changes in actinic fluxes. For example a 16 percent loss in stratospheric ozone resulted in a 39 percent increases in the tropospheric photolysis rate of ozone, with a corresponding increase in the NO_2 photolysis rate of less than 1 percent.

A base case simulation, conducted with the STEM-II/radiative code utilizing NAPAP emission inventories, and total ozone measurements from the Nimbus 7 satellite for June 1984, showed a strong correlation with ozone ground monitoring data. The simulation also captured the diurnal variations in ozone. During the episodic simulation, areas in the northeast experienced maximum ozone concentrations exceeding 175ppb.

A 16 percent reduction in total ozone, representative of stratospheric ozone conditions in the year 2004, decreased ground level ozone concentrations across the entire domain. This finding was the opposite of previous box model studies conducted by Liu and Trainer, in which ozone concentrations were shown to increase in areas with NO_X concentrations above 1ppb. These differences are attributed to transport processes, and highlight the importance of utilizing detailed models which include chemistry and transport mechanisms. Box model studies are strong indicators of the non-linear behavior of atmospheric chemistry, however, they should be used with caution when evaluating conditions influenced by transport of pollutants.

Ground level ozone concentrations were found to be sensitive to changes in NO_X emissions. A reduction in NO_X resulted in a decrease in ground level ozone concentrations across most of the domain. Exceptions occurred in areas with a low VOC/NOx concentration in which the effects on ground level ozone were minimal. With stratospheric ozone reductions and a decrease in NOx emissions, an enhanced ozone loss across the entire domain was experienced. Ozone concentrations were also reduced in the low VOC/NO_X corridors.

NO_X emission increases resulted in ground level ozone increases across the entire domain. Again, the exceptions were associated to areas with low VOC/NO_X ratio where the ozone concentrations did not reflect the changes in NO_X emissions. Overlaying stratospheric ozone loss and an NO_X emission increase, showed that for the northern regions of the domain, ozone concentrations were more sensitive to the 16 percent loss in overhead ozone, then to the 33 percent increase in NO_X emissions. The above resulted in ground level ozone concentrations decreasing across the northern sections of the domain. This area was dominated by high ozone levels for the base case simulation. The southern region, including the western edge of Florida through North Carolina experienced an increase in ground level ozone. This area is associated with high VOC/NO_X. Further details are presented in Crist et al., (1997).

An extensive study of the application of ADIFOR for sensitivity analysis of ozone and UVB using a coupled chemistry-radiation model was also conducted. The basic modeling approach is summarized in **Figure 2**. In this study a one-dimensional version of the STEM-II model was combined in an on-line manner with a 2-stream radiation model. The same models as discussed previously. This "system" was used to calculated the sensitivities of ozone concentrations and production/destruction rates, ozone photolysis rates, and UVB fluxes to changes in TOMS, meteorological factors (RH and temperature), NO_x and NMHC emissions, and aerosol properties (including aerosol composition and distribution). Sample results are presented in **Figures 2-5**, and in **Table 2**. Both rural and highly perturbed (urban) conditions were studied.

These studies revealed that ozone at these urban and rural locations showed a similar maximum sensitivity with respect to NO_x emissions. However at the urban sites, the near surface sensitivity is negative, and becomes positive above about 1 km. In contrast, ozone shows a positive sensitivity to hydrocarbon emissions throughout the profile. Near the surface the magnitudes of the sensitivities with respect to NO_x and NMHCs are similar, but at higher elevations the sensitivities with respect to NO_x are larger.

The sensitivity of ozone concentrations and photolysis rates are largest for changes in total ozone, and the sensitivities with respect to TOMS and aerosol properties are similar to those with respect to emissions. The overall effect of an increased aerosol loading is to decrease ozone photolysis rates and the surface UV-B flux. Increasing the relative humidity with regards to aerosol parameters could cause either an increase or decrease in ozone photolysis, depending on the type of aerosols considered. Furthermore, on a molecule for molecule basis, changes in tropospheric ozone have a larger effect on surface UV-B than changes in stratospheric ozone. Further details are presented in Hotchkiss (1996).

Linking of the STEM-II model with radiative transfer model on-line.

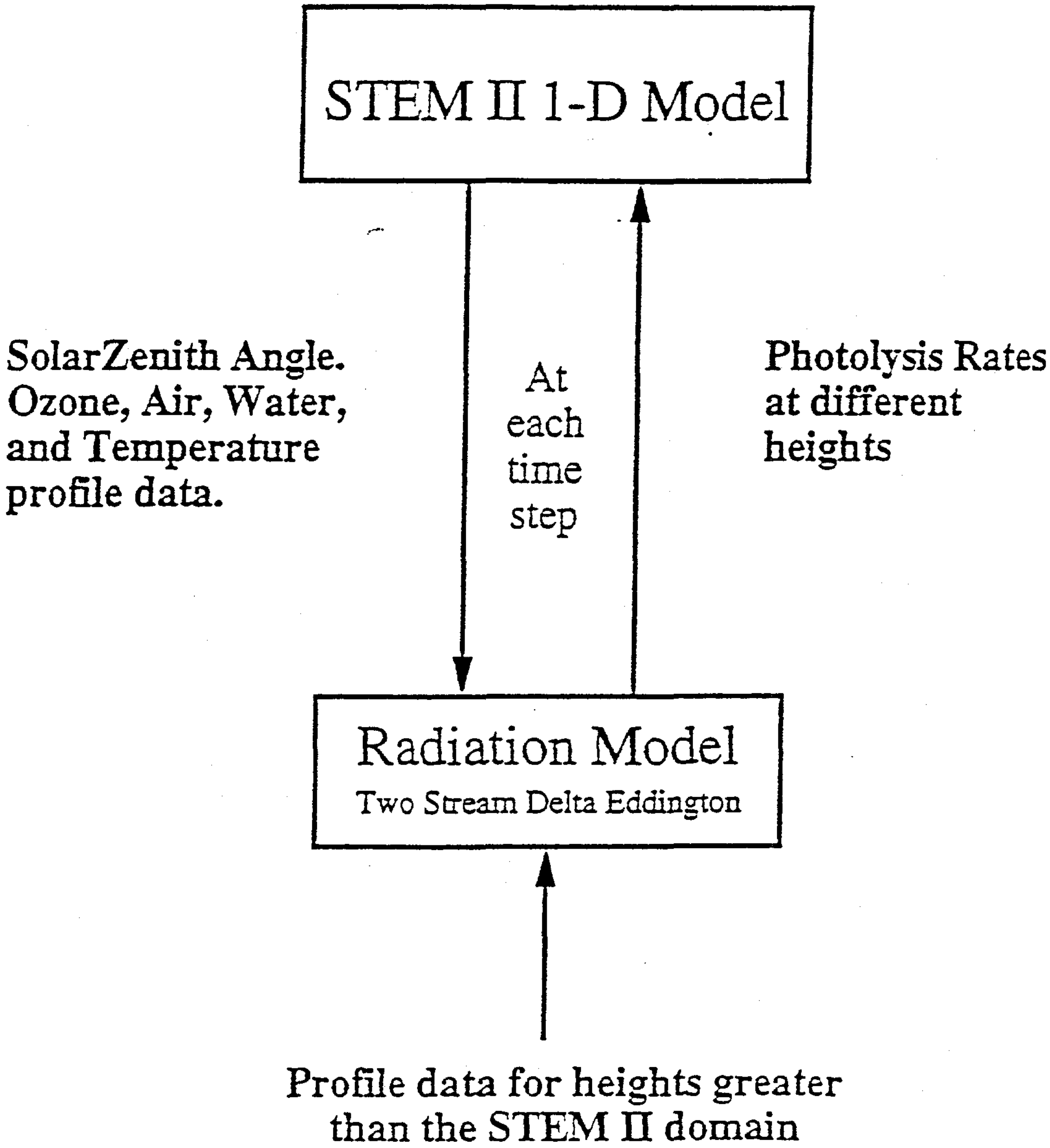
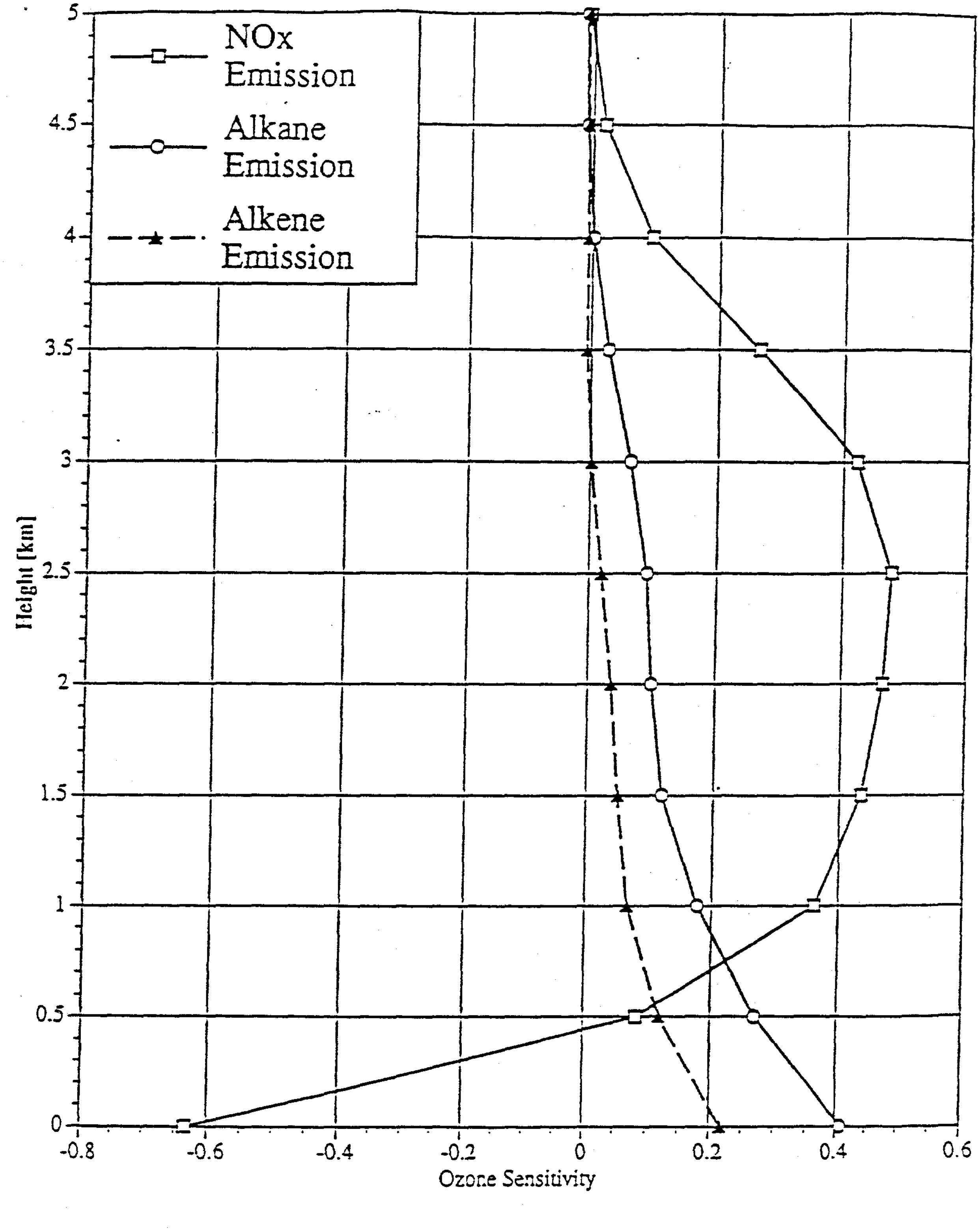
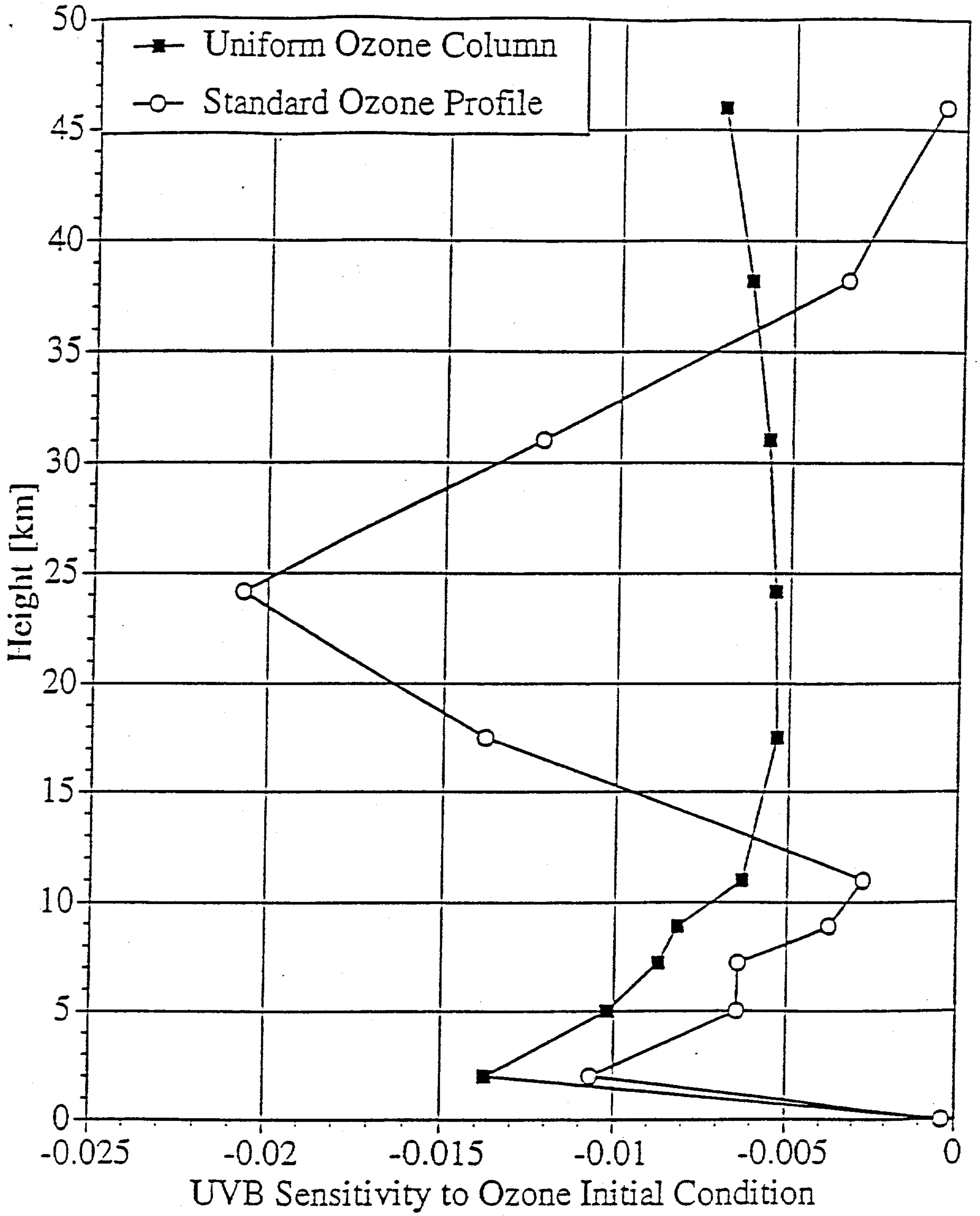


Figure 2. Combined chemistry-radiation model used in ADIFOR sensitivity analysis.



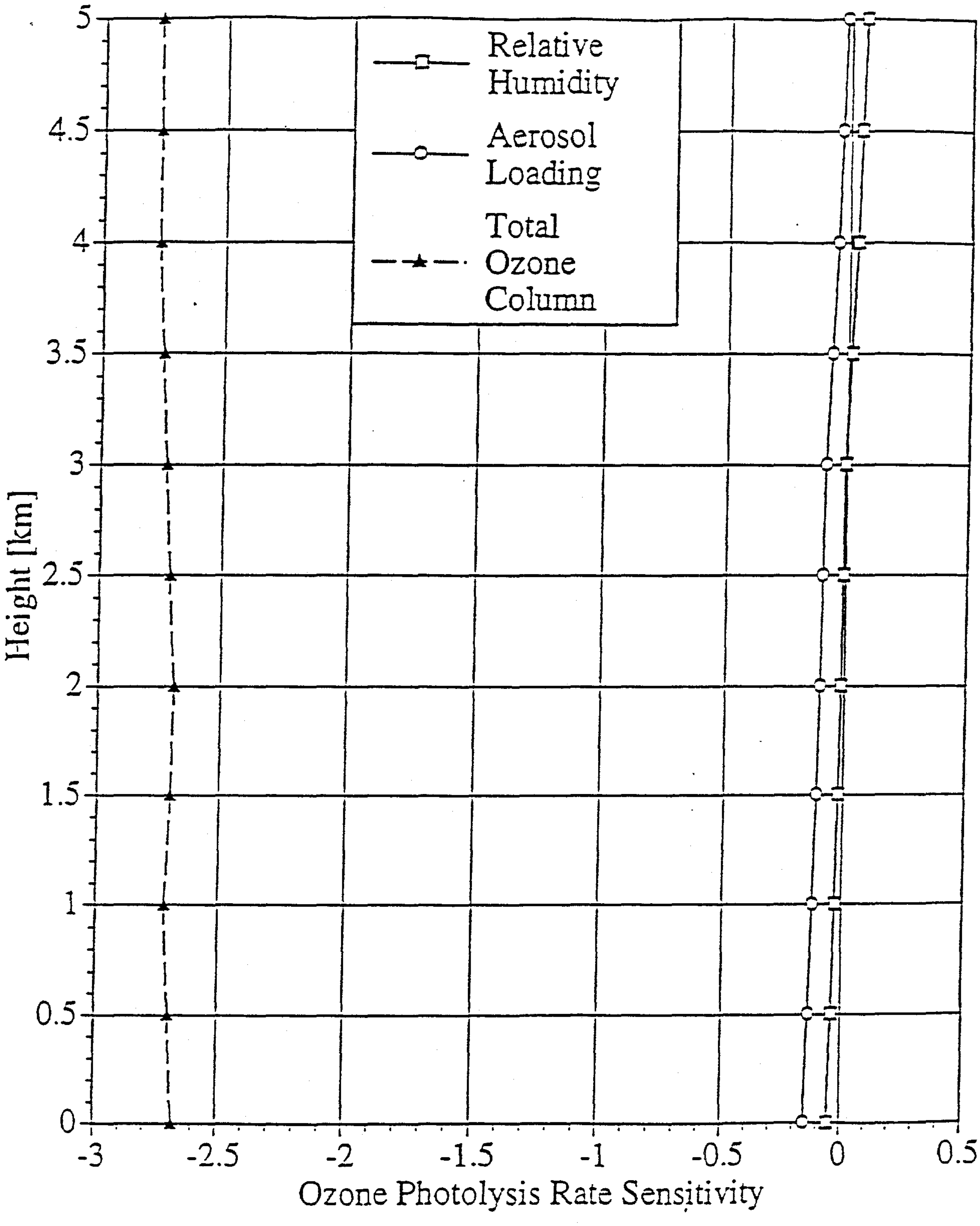
Ozone sensitivity to NOx, Alkanes (>C3), and Alkenes surface emissions.

Figure 3. Ozone sensitivity with respect to changes in surface emissions for the urban location.



Surface UV-B flux sensitivity to changes in ozone concentrations at certain heights.

Figure 4. Surface UVB sensitivities with respect to changes in ozone.



Ozone photolysis rate sensitivity to changes in radiation calculation inputs.

Figure 5. Ozone photolysis rate sensitivities with respect to aerosol loading, TOMS and relative humidity

Table 2. UV-B surface UVB sensitivities with respect to aerosol properties.

UV-B surface flux sensitvities for the different aerosol parameters and conditions.

	Connecticut	Connecticut^P	Georgia	Georgia^P
Aerosol Loading	-1.22e-1	-8.20e-1	-9.57e-2	-7.58e-1
Relative Humidity	-1.43e-2	-4.16e-1	-8.12e-2	4.82e-1
Ozone Column	-1.56		-1.30	

	Rural	Tropospheric[1]	Urban	Tropospheric[2]
Extinction	-2.94e-3	-1.84e-3	-3.27e-3	-2.03e-3
Absorption	-3.05e-3	-1.21e-3	-1.98e-2	-0.96e-3
Asymmetry Parameter	5.41e-3	7.58e-2	7.39e-3	7.09e-2

[P] Indicates constant aerosol profile

[1] Tropospheric aerosols with rural aerosols below them.

[2] Tropospheric aerosols with urban aerosols below them.

	Troposphere	Rural	Urban
Scattering Albedo , ϖ	0.9645	0.9517	0.7738
Asymmetry Parameter ,g	0.7176	0.7240	0.7713
Extinction Coefficient ,k_e [particle/cm^3]	1.8e-5	1.89e-5	1.96e-5

Aerosol type parameters (Shettle and Fenn, 1979) at 300 nm and 70% relative humidity.

The final studies of this project investigated aerosol impacts on the photochemical oxidant cycle; specifically photolysis rates and ozone production. Four representative aerosol types were examined (rural, urban, clean maritime, and maritime-mineral). The effect of relative humidity, the shape of the vertical profile and emissions rates were studied. The presence of aerosols in the boundary layer is found to enhance or inhibit ozone formation depending on the optical properties of the particles. Ozone production in the mid-troposphere is usually enhanced due to backscattering by the boundary layer aerosol. NO2 photolysis rates and ozone production were found to be most sensitive to the presence of strongly absorbing aerosol. The sensitivity of photolysis rates and ozone production were found to be as sensitive to relative humidity effects as to aerosol loading. Further details are presented in the attached papers.— Reprint removed for separate processing

3. SUMMARY

Much has been accomplished during the project period. The radiative transfer and STEM-II models have been linked and used to investigate UVR-chemistry interactions. Specifically studies on the effect of changes in atmospheric ozone on UVB surface fluxes and the feedback between changes in UVB fluxes and tropospheric photochemistry have been performed. Results have identified many of the important feedbacks, and shown that multidimensional models are needed to capture these interactions. We have also successfully implemented ADIFOR with our model and obtained sensitivities of ozone with respect to initial conditions and rate constants. We have demonstrated that ADIFOR can be used for these applications and are excited by the prospects offered by ADIFOR. We also have developed new ideas regarding how ADIFOR can best be exploited for these applications. Substantial progress has also been made in the area of improving the computational efficiency of the models.

4. Deliverables

Publications:

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