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Development and Testing of an Improved Photolysis Rate Model for Regional Photochemical Modeling

Shawn J. Roselle*, Jonathan E. Pleim*, Kenneth L. Schere*

Atmospheric Sciences Modeling Division, Air Resources Laboratory, National Oceanic and Atmospheric Administration, Research Triangle Park, North Carolina 27711

*On assignment to the National Exposure Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.

Adel F. Hanna, and Ji-Cheng C. Jang

MCNC-North Carolina Supercomputing Center, Research Triangle Park, North Carolina 27709

Introduction

Almost all chemical reactions in the atmosphere are initiated by the photodissociation of a number of trace gases. These reactions are responsible for most of the buildup of smog, which has detrimental effects on human, animal, and plant life. In order to accurately model and predict the effects of air pollution, good estimates must be made of the rates of these photochemical reactions. A direct measure of this photodissociation is the photolysis rate. Many current air quality models use crude approximations to calculate the photolysis rates with little consideration of variations in vertical profiles of temperature, ozone concentrations, aerosol concentrations, cloud parameters, and spectral surface albedo. However, more accurate estimates of photolysis rates can be produced with advanced radiative transfer modeling techniques and with the utilization of detailed measurements.

An advanced computational model has been developed to simulate the actinic flux and photolysis rates of tropospheric species for air quality and tropospheric photochemical modeling. The model combines advanced radiative transfer models with explicit computations of photolysis rates using detailed information on prevailing atmospheric conditions during simulation episodes, satellite ozone data, and detailed characteristics of clouds, aerosols and surface albedo. In this paper we discuss the advantages of using the refined model to simulate photolysis rates over a standard lookup table method. We also discuss the sensitivity of the photolysis rate simulations to the radiative transfer scheme used as well as key parameters such as cloud, ozone profiles and surface albedo.

Background

Photodissociation is the conversion of solar radiation into chemical energy to activate and dissociate chemical species. Examples of species that photodissociate include many important trace constituents of the troposphere such as NO_2, O_3, HCHO, CH_3CHO, HONO, the NO_3 radical, and H_2O_2. The accuracy of the simulation of the entire chemical system is highly dependent upon the

accuracy of the rates of photolysis which are the primary source of radicals to the system. Photolysis rates (min^{-1}), also called j-values, are computed as

$$J = \int_{\lambda_1}^{\lambda_2} F_\lambda \sigma_\lambda \phi_\lambda d\lambda$$

where, F_λ is the actinic flux (photons cm^{-2} min^{-1}), σ_λ the absorption cross section (cm^2 $molecule^{-1}$), ϕ_λ the quantum yield (molecules $photon^{-1}$), and λ the wavelength (nm). Absorption cross sections and quantum yields are molecular properties that are functions of wavelength and temperature and are unique to each species. Laboratory experiments have been conducted for many of the species that photodissociate in the troposphere to measure the absorption cross sections and quantum yields. Actinic flux, on the other hand, is a radiometric quantity that is a measure of the integrated spectral radiance over all solid angles per unit area. The spherical receiving surface distinguishes the actinic flux from the more commonly measured irradiance, which is the radiance falling on a horizontal surface. Thus, the actinic flux can be called spherical spectral irradiance. The actinic flux changes with time of day, longitude, latitude, and season, and is governed by the astronomical and geometrical relationships between the sun and the earth; it is greatly affected by the earth's surface as well as by various atmospheric scatterers and absorbers. All of these factors make quantifying atmospheric radiative transfer very complicated. Hence, correct model calculation of the temporal and spatial variation of the actinic flux is critical to obtaining accurate photolysis rates for regional and mesoscale episodic photochemical modeling.

Model Description

An advanced model (JPROC: J-value PROCessor) has been developed to calculate photolytic rate constants for regional air quality models. JPROC is largely based on the Madronich's model for Tropospheric Ultraviolet and Visible (TUV) radiation[1-2]. JPROC adds to TUV the ability to calculate photolysis rates for all grid cells of a specified Eulerian air quality model. In our case, we are applying JPROC to EPA's Models-3 Community Multiscale Air Quality Model (CMAQ)[3]. Other features that have been added to TUV include the ability to compute photolysis rates for each simulation hour, making use of datasets available from the Mesoscale Meteorological Model version 5 (MM5)[4] and from other available sources. JPROC is flexible in the specification of wavelength bands, extraterrestrial irradiance data, ozone profiles, aerosol profiles, cloud distributions, and absorption cross section and quantum yield data.

Radiative Transfer

Radiative transfer models, including two-stream and multi-stream discrete ordinate approximations[2], are used for computing the actinic flux. Two-stream models make two basic assumptions: (1) the phase function is completely isotropic; and (2) radiances in the upward and downward hemispheres are individually isotropic. The two-stream approximations are limited in application to cases where the scatter is not highly anisotropic. The multi-stream radiative transfer models divide the radiation field into more than the two described above, and include all orders of multiple scattering. This is particularly important when considering radiative effects of clouds and aerosols. JPROC uses these

models to provide detailed descriptions of the radiative processes in the atmosphere; detailed descriptions of clouds, aerosols, ozone absorption, oxygen absorption in the Schuman-Runge Bands[5], Rayleigh scattering[6] and surface albedo are provided to the radiation models. One of the most critical elements in calculating the actinic flux is to describe the structure of the atmosphere as accurately as possible, making use of as much data as possible on the episodic meteorological and chemical conditions.

Vertical Coordinate

The vertical resolution for calculating photolysis rates in JPROC (i.e. the vertical grid and number of layers) is set to correspond with the air quality model's vertical coordinate system. In the case of Models-3/CMAQ, a terrain-following cordinate system is used in specifying the vertical model structure. Depending on the model application, there may be any number of vertical layers, although typically the troposphere is subdivided into 6 to 30 layers. Since the radiative transfer calculations must consider the effects of the stratosphere, the vertical coordinate for JPROC extends above that of CMAQ, through most of the stratosphere, to a height of 50 km.

Surface Albedo

An important parameter for radiative transfer calculations is the earth's surface albedo. The albedo data given by Demerjian et al.[7] have been used extensively in radiative transfer models. These data are given as a function of wavelength, but not of landuse type. Current air quality modeling domains cover most of the United States, including a broad range of land surface types. Therefore, more detailed spatial data are needed to fully resolve the details within modeling domains. A good candidate for JPROC is MM5's spatially-resolved surface albedo data with solar zenith angle variance. However, MM5's data does not have the wavelength distribution needed for the radiative transfer calculations. Therefore, a procedure was developed in JPROC to make use of both datasets to resolve the spectral and spatial distributions of the surface albedo. MM5's surface albedo data are treated as broad-band integrated values, which get distributed across wavelengths using a normalized profile of the albedo data from Demerjian et al.[7].

Temperature Profile

Several factors in JPROC depend on the temperature profile, including ozone absorption, SO_2 absorption, and the absorption cross sections and quantum yields for individual photolysis reactions. To improve the accuracy of these calculations, spatially variant temperature profiles from MM5 are used in JPROC to describe the temperature profiles within the troposphere. Above the tropopause, JPROC uses the U.S. Standard Atmosphere[8] temperature profile.

Aerosols

The radiation field, especially in the atmospheric boundary layer, is greatly affected by aerosols. The important aerosol properties that affect the atmospheric radiative transfer are size distribution, number density, and complex refractive index. Currently, a single profile of aerosol number density[9] is being used for all grid cells in JPROC. The utility of using dynamically-calculated aerosol profiles, as simulated by the Models-3 particulate model[10] will be investigated in the future as an input to JPROC. Such an approach may result in considerable improvements in the accuracy of photolysis rates especially during regional haze episodes in the U.S.[11].

Ozone Absorption

Absorption by ozone is calculated using the most recent NASA recommendations[12]. Ozone profiles are set by taking the U.S. Standard Atmosphere[8] O_3 profile and uniformly rescaling this profile for all model grid cells to match the integrated total ozone column as measured by the Total Ozone Mapping Spectrometer (TOMS) instrument aboard the sun-synchronous polar orbiting Nimbus 7 satellite. TOMS data (currently Version 7) are archived and available at the National Satellite Service Data Center (NSSDC) in the form of digital daily maps with a resolution of 1 degree latitude by 1.25 degrees longitude, for the satellite's operating period November 1978 - May 1993. The TOMS data provide a daily measurement of the total column ozone as gridded information that feeds into the radiative transfer model to calculate the actinic flux. Bilinear interpolation is used to map the TOMS data to the model grid system. In the absence of TOMS data, the U.S. Standard Atmosphere[8] ozone profile is used.

Cloud Droplet Scattering and Absorption

Meteorological modeling includes the use of sophisticated cloud parameterization schemes[13] to provide pertinent physical characteristics of clouds to air quality models. These schemes provide detailed descriptions of cloud dynamics and microphysics that are necessary for chemical and radiative transfer parameter calculations. The radiative transfer model uses detailed cloud parameters such as the profiles of the liquid water content and droplet size distribution to calculate actinic fluxes.

Cloud coverage is a critical parameter for calculating actinic fluxes and photolysis rates. JPROC takes cloud cover information to determine if the radiative transfer calculations will be for either clear or cloudy sky conditions. For grid cells with completely clear skies, only one radiative transfer calculation is performed using clear sky conditions. For overcast grid cells, again only one radiative transfer calculation is made using the cloudy sky conditions. For cells with fractional cloud coverage, the radiation model is run twice, once for the clear sky fraction and another time for the cloudy fraction. A weighted average is then performed on the actinic flux[14] to determine a grid cell average value.

Species Kinetic Parameters

Periodic updates of the absorption cross sections and quantum yields for different species are published by different organizations[12]. JPROC provides the capability of computing photolysis rates for any chemical mechanism, using absorption cross section and quantum yield data specified by the user. Default sets have been set up for the Carbon Bond Mechanism IV[15], the Regional Acid Deposition Model mechanism version 2 (RADM2)[16], and the SAPRC mechanism[17]. In addition, users can deviate from these standard sets to test other data, including the revisions suggested by NASA[12].

Results

Table lookup

One method being used to specify photolysis rates for air quality models involves the calculation of rates for various predefined conditions, such as different zenith angles, altitudes, albedos, total ozone

column values, etc., and the interpolation of photolysis rates from this predescribed table using the current conditions (e.g. current zenith angle, etc). This method has been employed in the Regional Acid Deposition Model (RADM)[18]. We will use RADM's approach as the basis for comparison. RADM's table has three dimensions of interpolation, including the hour from local solar noon, latitudinal band, and altitude. Values in the table have been computed using the Delta-Eddington two-stream method[19] for calculating the actinic flux. In addition, the vertical profiles of temperature, air number density, and ozone vary latitudinally and are interpolated from seasonal data. Within the RADM, values are interpolated from the table to each model grid cell and layer, and then a correction is applied to the interpolated value to take into account the effects of clouds. In this application, there were 33 columns, 36 rows, and 6 layers in the modeling domain, with a horizontal grid size of 80 km.

Figure 1 shows a spatial plot of the interpolated table photolysis rates for NO_2 on August 2, 1988 at 18:00Z for an 80 m layer just above the earth's surface (model layer 1), which is near local noon and the daily maximum for photolysis rates (j-values). The photolysis rates for NO_2 (or $J(NO_2)$ values) are high across much of the modeling domain, with values ranging from 0.520 min^{-1} to 0.570 min^{-1}. A few locations have higher values, but these are very limited in spatial extent. Several areas have lower values (denoted by the darker shading), particularly the "bullseyes" over Mississippi/Alabama and Georgia/Tennessee, as well as the broad band across most of Ontario, Canada. All of these areas have cloud cover which is attenuating the actinic flux and lowering the photolysis rates. Figure 2 shows the percentage cloud cover over the modeling domain.

Figure 3 shows a vertical cross section of j(NO2) corresponding to the same hour as Figure 1. Here, we have selected an x-z cross-section through northern Georgia. Higher rates in the upper model layers (e.g. layers 5 and 6) appear over columns 16-25. This is the location of the highest cloud cover fraction and shows the enhancement of photolysis rates by reflections above the cloud top. Through the cloud layer, the rates decrease linearly to the cloud base. Below the cloud, the photolysis rates are much lower than surrounding grid cells.

JPROC

Comparison to Table lookup plots

Our first simulation with the JPROC model uses a two-stream radiative transfer model, and incorporates TOMS and MM5 data. The same cloud inputs (i.e. cloud cover, cloud base, cloud top, and liquid water content) and chemical data (e.g. cross sections and quantum yields) were used in JPROC as were used in the interpolated table simulation. Results were directly compared with the RADM interpolated table results. Similar comparisons were conducted by Hass and Ruggaber[20]. Figure 4 shows the percent difference in $j(NO_2)$ between the interpolated table values and the JPROC calculated values for model layer 1 at 18:00Z on August 2, 1988. The values are similar across the Mid-Western and Atlantic Coastline States, while values are lower for JPROC over portions of Georgia, Tennessee, Alabama, Mississippi, Ontario, Canada, and the open waters of the Gulf of Mexico and Atlantic Ocean. On average, $j(NO_2)$ for JPROC is 4% lower than for the RADM interpolated table values, with the largest difference of 70% occuring over northern Georgia.

Looking next at the percent difference plot in $j(NO_2)$ (Figure 5) along the same X-Z cross section plot as Figure 3 shows that the differences are small except in the vicinity of clouds. Values are lower for JPROC above some clouds, but are higher by as much as 36% above the clouds in

northern Georgia. Under the same cloud mass, we find that the NO_2 photolysis rates are lower for JPROC by as much as 42%. This illustrates that the treatment of clouds can have a significant impact on calculated photolysis rates. For JPROC, the cloud parameters are input before the radiative transfer calculations, whereas for the RADM interpolated table the cloud parameters are used to adjust precalculated clear sky j-values.

Two-stream vs. Multi-stream

Next, we replaced the two-stream radiation model with a multi-stream discrete ordinates model in JPROC. The multi-stream code is quite complex and consumed about 150 times more computer CPU time than the two-stream code. Because of this, we wanted to check whether the two-stream code results differed significantly from those of the multi-stream code. Zeng et al.[2] have previously compared relative changes in j-values for various cloud optical depths using the same two-stream and multi-stream models. They found that the two-stream model overestimates j-values by 2-10% under the cloud layer. Figure 6 shows the percent difference in $j(NO_2)$ for our simulations with JPROC. Over most of the modeling domain, the differences are small between the two radiation models, with an average difference of less than 1%. Over the northern regions, the multi-stream model gives slightly higher photolysis rates (less than 5%), while over the Gulf of Mexico and Atlantic Ocean the two-stream model gives higher photolysis rates (by at most 13%). A vertical cross section plot (not shown) of the differences showed that the multi-stream model led to higher photolysis rates in the upper model layers.

TOMS data comparison

Another input to the model that can be improved upon is the specification of the total ozone column data. We have compared two runs of JPROC, one using the U.S. Standard Atmosphere[8] profile for ozone and the other with the same profile rescaled to the total ozone column as specified in the TOMS database for each grid cell. Differences in $j(NO_2)$ between the two simulations were examined first, but were found to be very small (< 0.5%). This was expected because most of the ozone absorption occurs in a different wavelength band than the band for NO_2 photolysis. Therefore, we looked at the O3->O(1D) photolysis reaction to see the effects that TOMS data can have on other photolysis rates. The percent difference in the O_3->$O(^1D)$ photolysis reaction rate is shown in Figure 7. Differences are highest in the southern part of the modeling domain, where the simulation with TOMS data had photolysis rates over 36% higher than the simulation with the standard profile. Photolysis rates were about 20% higher in the TOMS run over most of the modeling domain. Almost every grid cell in the modeling domain had lower total ozone column values than the 320DU value of the U.S. Standard Atmosphere. The average total ozone column in the modeled region was 305 DU. The results shown by these two simulations are expected to be highly variable since the total ozone column values have considerable seasonal and latitudinal variation. Our result agrees with the results of Hass and Ruggaber[20] demonstrating that the use of TOMS data can better resolve episodic estimates of photolysis rates.

Albedo sensitivity

Another tested sensitivity was the specification of surface albedo. We ran JPROC with an albedo of 0.1 for all grid cells and then a second time with MM5's spatially varying albedo data. The percent difference plot in $j(NO_2)$ is shown in Figure 8. Little differences are noted over the water areas where MM5's albedo is close to 0.1. Larger differences are noted over all land areas, where the

MM5 data led to more than a 5% increase in NO_2 photolysis rates. Over the entire domain, $j(NO_2)$ was 3% higher in the simulation with MM5 albedo data.

Conclusions

An advanced photolysis rate model was developed for use in air quality models. The model (JPROC) incorporates information on modeled meteorological profiles and cloud cover, and total ozone column data, and provides considerable improvements over current methods used to compute photolysis rates. JPROC has been tested in EPA's Models-3/CMAQ. The calculated photolysis rates were similar to rates previously used in the Regional Acid Deposition Model (RADM), with most of the differences occurring immediately above and below clouds. Differences in photolysis rates using either a two-stream or a multi-stream radiative transfer model were smaller than the differences between the 3-dimensional two-stream model and the lookup table approach used in the RADM. Results also showed that the specification of total ozone column can significantly impact the rates for some photolysis reactions. Another sensitivity test was conducted on the use of MM5's spatially-varying albedo data; over most land areas the NO_2 photolysis rate increased by 5-7% when MM5's albedo was used compared to simulations using a spatially uniform albedo of 0.1.

Much work is still needed to fully test the results of JPROC. First and foremost, the results must be evaluated against any available datasets. Second, we are in the process of testing different treatments of clouds, including methods to link JPROC with MM5, and the utility of satellite cloud coverage data[21]. Another important area to be improved upon is in the area of aerosols. Models-3 will have detailed aerosol distribution information, and we should be able to utilize this data to improve the photolysis rate calculations within the boundary layer.

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We are very grateful to Sasha Madronich for sharing his TUV model with us, upon which our model is based. This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

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Figure 1. NO_2 photolysis rates (min^{-1}) for layer 1 on August 2, 1988 at 1800Z, interpolated from the RADM table with cloud cover corrections applied.

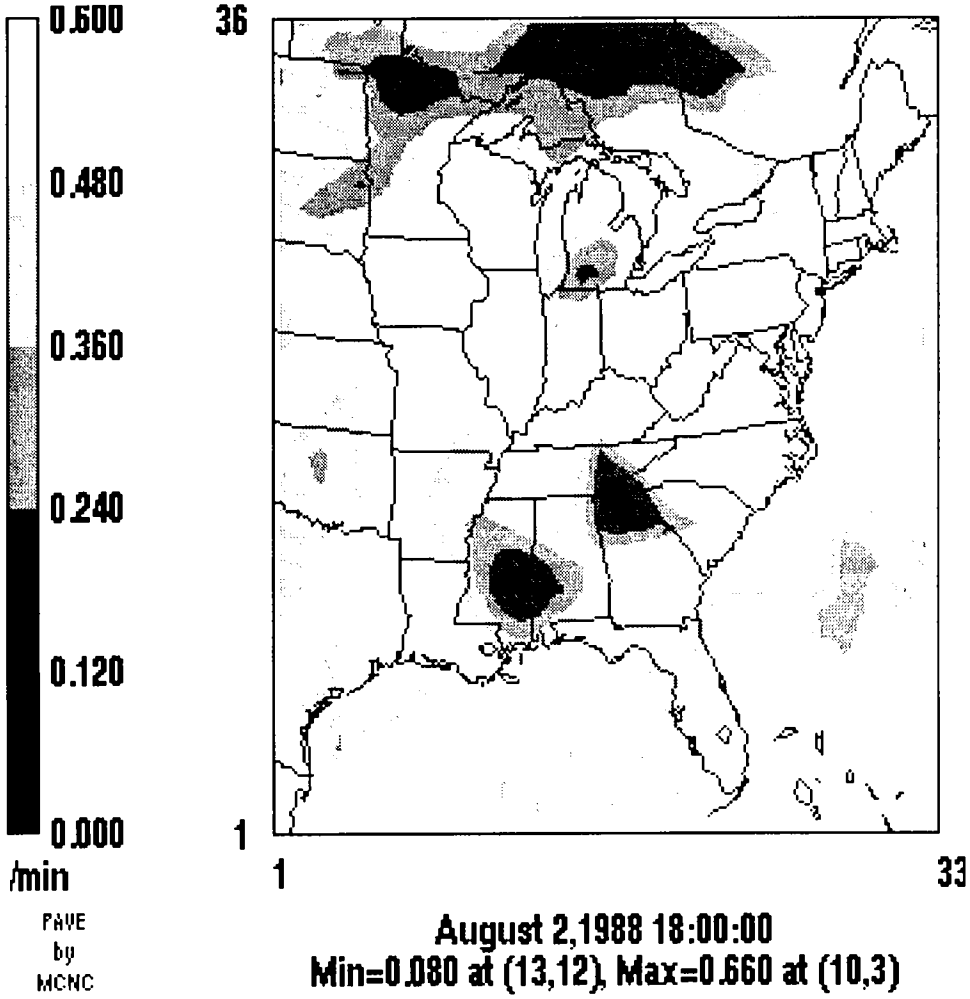


Figure 2. Cloud cover fraction used in RADM on August 2, 1988 at 1800Z.

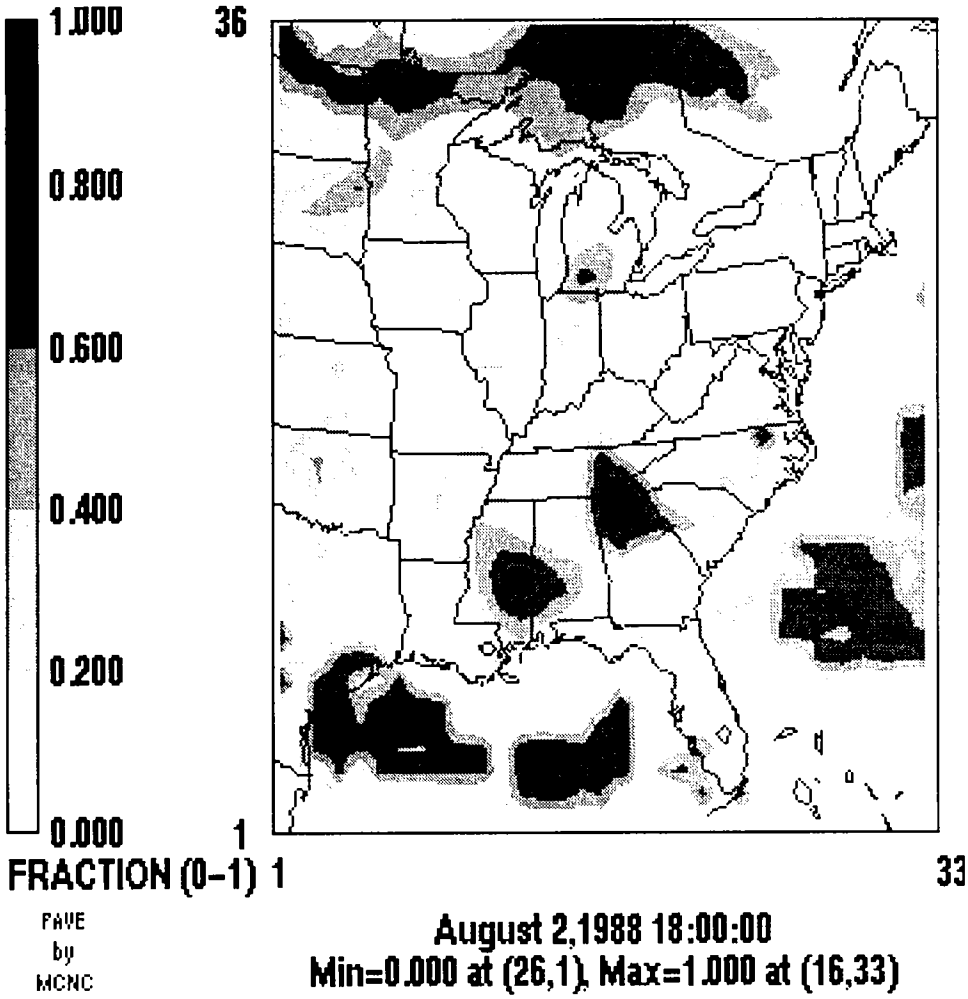


Figure 3. Vertical cross section of the NO_2 photolysis rates (min^{-1}) on August 2, 1988 at 1800Z, interpolated from the RADM table with cloud cover corrections applied. Vertical slice through cloud mass over northern Georgia.

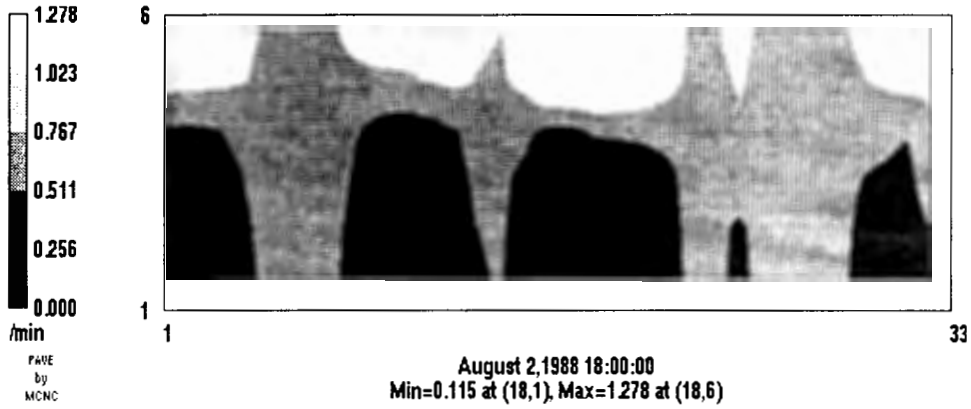


Figure 4. Percent difference in NO_2 photolysis rates on August 2, 1988 at 1800Z for layer 1 between the interpolated RADM table and the JPROC calculated values.

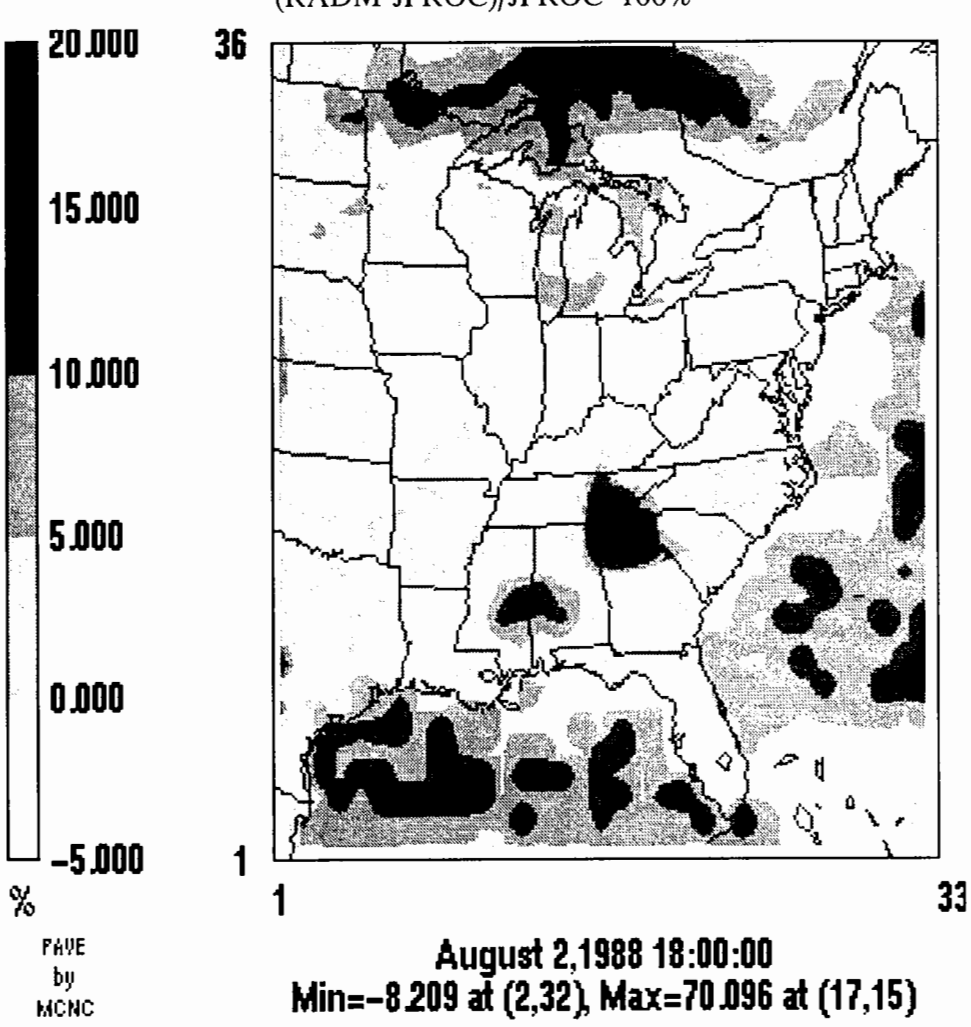


Figure 5. Vertical cross section of the percent difference in NO_2 photolysis rates for layer 1 on August 2, 1988 at 1800Z between the interpolated RADM table and the JPROC calculated values.

(RADM-JPROC)/JPROC*100%

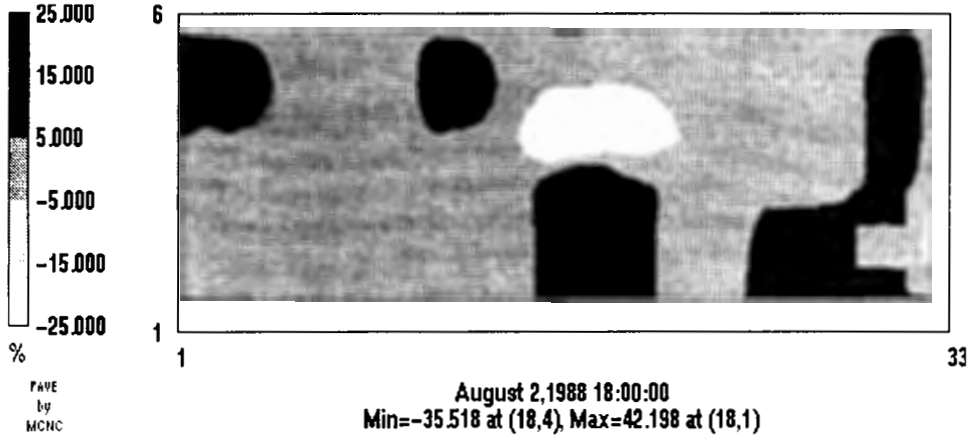


Figure 6. Percent difference in the NO_2 photolysis rates on August 2, 1988 at 1800Z for layer 1 between the JPROC two-stream calculations and the multi-stream calculations.

(TWOstr-MULTstr)/MULTstr*100%

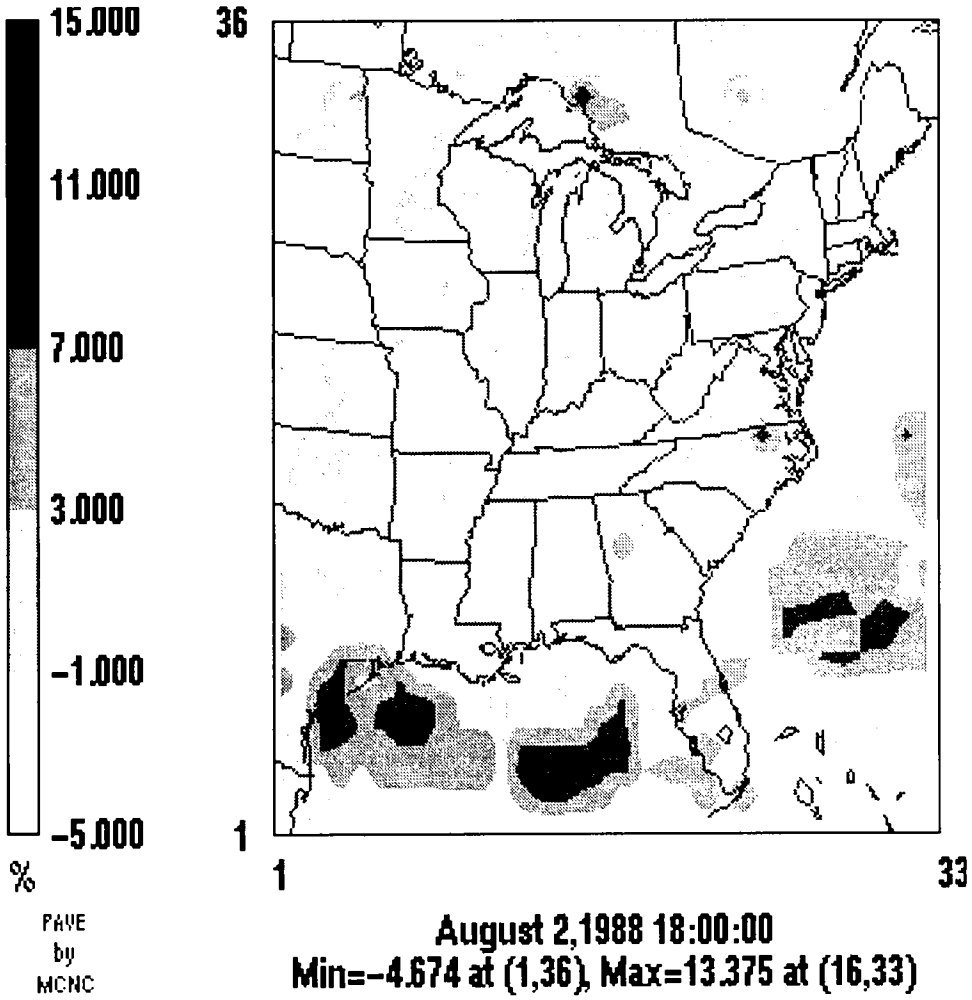


Figure 7. Percent difference in the O_3->$O(^1D)$ photolysis rates on August 2, 1988 at 1800Z for layer 1 between the JPROC calculations with TOMS data and with the U.S. Standard Atmosphere ozone profile.
(TOMS-STDO3)/STDO3*100%

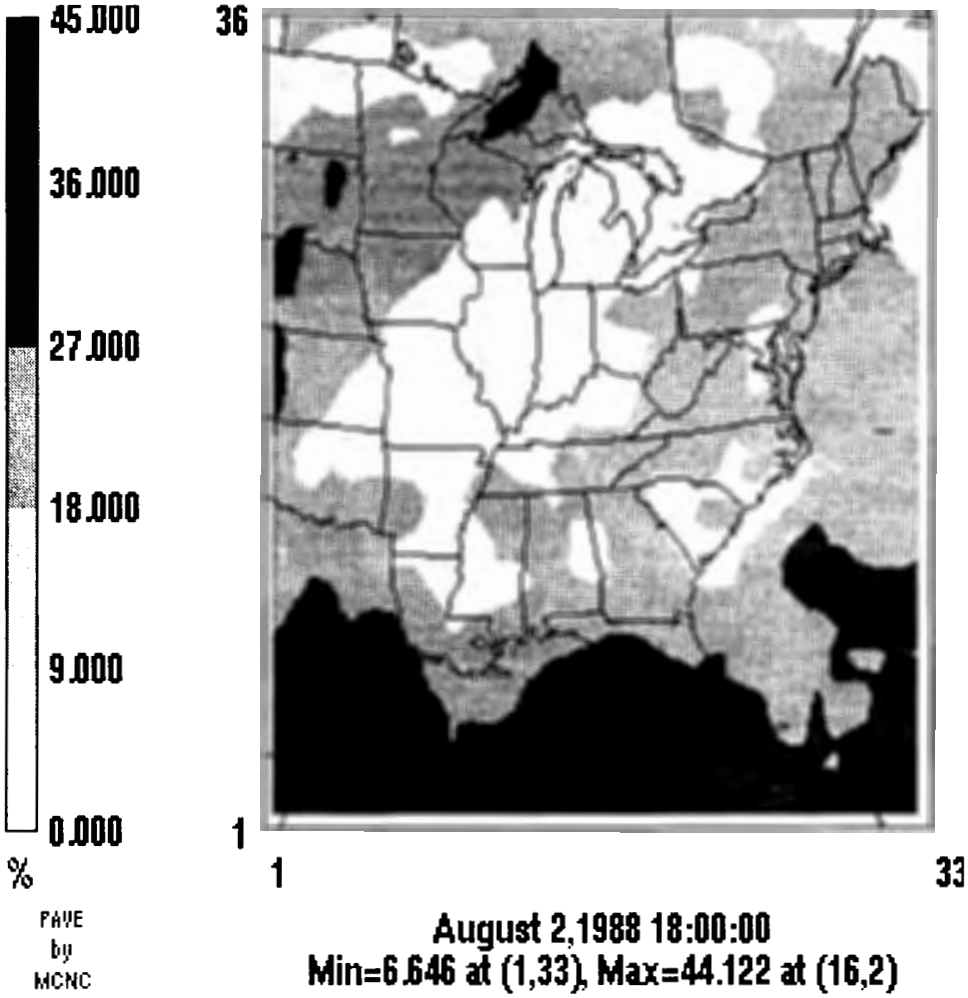
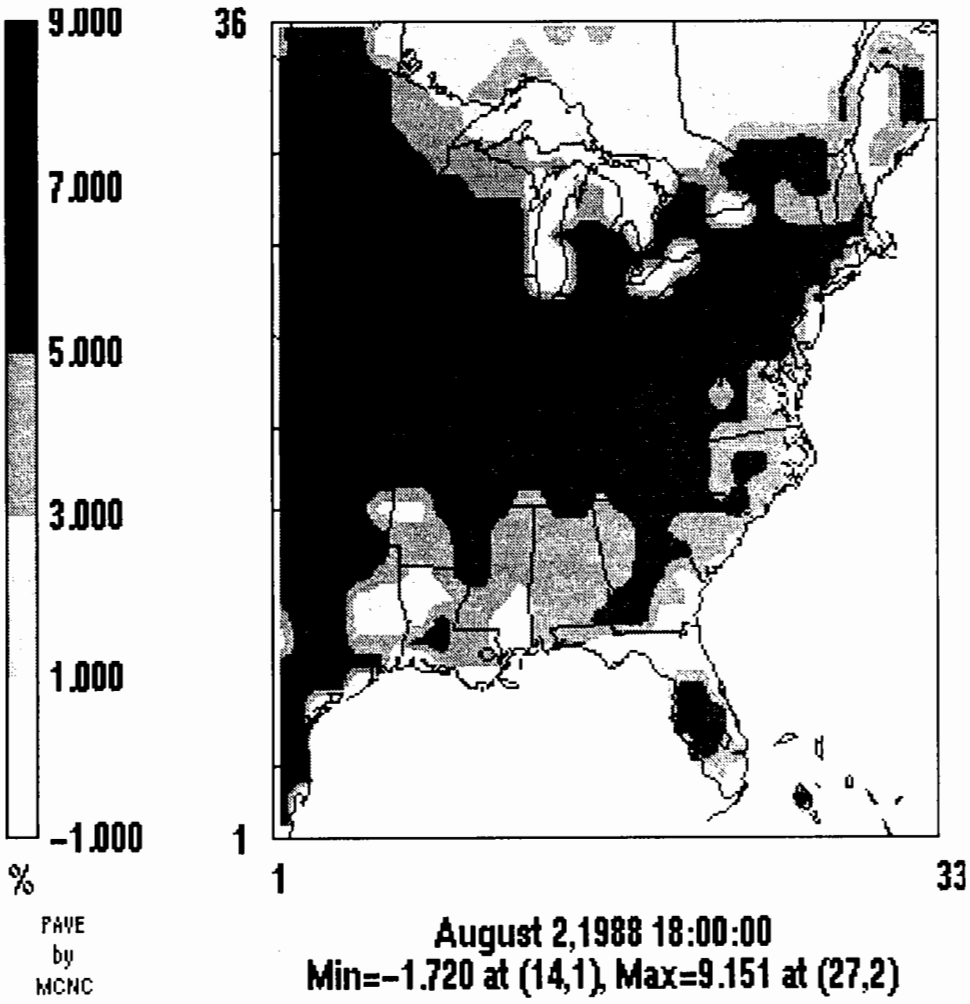


Figure 8. Percent difference in the NO_2 photolysis rates on August 2, 1988 at 1800Z for layer 1 between the JPROC calculations with MM5's albedo data and with spatially uniform albedo data.

(MM5alb-UNIFalb)/UNIFalb*100%



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16. ABSTRACT

An advanced computational model has been developed to simulate actinic flux and photolysis rates of chemical species for air quality and tropospheric photochemistry modeling. The module combines advanced radiative transfer models with explicit computations of photolysis rates using detailed information on prevailing atmospheric conditions during simulation episodes, satellite ozone data, and characteristics of clouds, aerosols and surface albedo on a time dependent basis over a 3-dimensional grid. The sensitivity of the photolysis rate simulations to different radiative transfer schemes as well as key parameters such as ozone profiles and surface albedo is examined. Photolysis rate results are similar to rates previously used in the Regional Acid Deposition Model (RADM), with most of the differences occurring immediately above and below clouds. Differences in photolysis rates using either a two-stream or a multi-stream radiative transfer model were smaller than the differences between the 3-dimensional two-stream model and the lookup table approach use in the RADM. Results also showed that the specification of total ozone column can significantly impact the rates for some photolysis reactions; in our test case, using TOMS data to specify the total ozone column for the 3-dimension grid increases the O3->O(1D) photolysis rate by ~20% over most of the modeling domain compared to a simulation using the U.S Standard Atmosphere ozone profile. Our last sensitivity test was the use of MM5's spatially-varying albedo data; over most land areas the NO2 photolysis rate increased by 5-7% when MM5's albedo is used compared to simulations using a spatially uniform albedo of 0.1.

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