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Correcting photolysis rates on the basis of satellite observed clouds

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[1] Clouds can significantly affect photochemical activities in the boundary layer by altering radiation intensity, and therefore their correct specification in the air quality models is of outmost importance. In this study we introduce a technique for using the satellite observed clouds to correct photolysis rates in photochemical models. This technique was implemented in EPA's Community Multiscale Air Quality modeling system (CMAQ) and was tested over a 10 day period in August 2000 that coincided with the Texas Air Quality Study (TexAQS). The simulations were performed at 4 and 12 km grid size domains over Texas, extending east to Mississippi, for the period of 24 to 31 August 2000. The results clearly indicate that inaccurate cloud prediction in the model can significantly alter the predicted atmospheric chemical composition within the boundary layer and exaggerate or underpredict ozone concentration. Cloud impact is acute and more pronounced over the emission source regions and can lead to large errors in the model predictions of ozone and its by-products. At some locations the errors in ozone concentration reached as high as 60 ppb which was mostly corrected by the use of our technique. Clouds also increased the lifetime of ozone precursors leading to their transport out of the source regions and causing further ozone production down-wind. Longer lifetime for nitrogen oxides (NO_x = NO + NO_2) and its transport over regions high in biogenic hydrocarbon emissions (in the eastern part of the domain) led to increased ozone production that was missing in the control simulation. Over Houston-Galveston Bay area, the presence of clouds altered the chemical composition of the atmosphere and reduced the net surface removal of reactive nitrogen compounds. Use of satellite observed clouds significantly improved model predictions in areas impacted by clouds. Errors arising from an inconsistency in the cloud fields can impact the performance of photochemical models used for case studies as well as for air quality forecasting. Air quality forecast models often use the model results from the previous forecast (or some adjusted form of it) for the initialization of the new forecast. Therefore such errors can propagate into the future forecasts, and the use of observed clouds in the preparation of initial concentrations for air quality forecasting could be beneficial.

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1. Introduction

[2] A key component of air quality modeling is the correct estimation of photodissociation reaction rates (or photolysis rates) for chemical species. These rates (the rate at which photochemistry takes place) depend on the intensity of solar radiation reaching a given point in the atmosphere and the molecular properties of the molecule undergoing photodissociation. Therefore attenuation or enhancement of radiant energy due to atmospheric absorption and scattering is an important factor in determining the photolysis rates. Since

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clouds can significantly alter the solar radiation in the wavelengths affecting the photolysis rates, they can have considerable impact on the photochemistry.

[3] Reliable estimates of photolysis rates are essential in reducing the uncertainty in air quality modeling. Air quality models rely on radiative transfer models for the prediction of photolysis rates. There are a suite of radiative transfer models [see *Barker et al.*, 2003] that take extraterrestrial solar flux, optical properties of the atmosphere, and surface albedo as input to describe the propagation of radiation in the atmosphere. These models are widely used for both research and in weather and climate models. *Barker et al.* [2003] compared the performance of 25 radiative transfer models with respect to the impact of unresolved clouds; most of the models used in their study underestimated atmospheric absorption of solar radiation. Other studies [*Collins et al.*, 2000; *Liao et al.*, 1999; *Jacobson*, 1998; *Dickerson et al.*, 1997; *Castro et al.*, 1997; *Ruggaber et al.*, 1994; *Madronich*, 1987] have investigated the effects of changes in atmospheric conditions and surface albedo on the estimates of photolysis rates. Most of these studies conclude that aerosols and clouds play an important role in modifying the photolysis rate either by enhancing it because of light scattering, or by reducing it because of absorption and attenuation of light.

[4] The Community Multiscale Air Quality modeling system (CMAQ) [*Environmental Protection Agency* (*EPA*), 1999] uses a two step approach for calculating the photolysis rates. This approach is similar to that of the Regional Acid Deposition Model (RADM) [*Chang et al.*, 1987] and is a typical method used in most air quality models. First, in a preprocessor, a radiative transfer module (based on *Madronich* [1987]) is used to compute clear sky photolysis rates for a range of latitudes, altitudes, and zenith angles. Then, within the chemical transport model, the tabular photolysis rates are interpolated for each location and corrected for cloud cover. There are two major concerns with this approach as far as cloud correction is concerned. First, estimation of cloud transmissivity in models is highly parameterized and, therefore, introduces a large uncertainty. Second and most important, the cloud information is provided by a mesoscale model, which has difficulty with the spatial and temporal placement of clouds and their vertical extent. The mesoscale model used in the CMAQ modeling system is the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5) [*Grell et al.*, 1994; *National Center for Atmospheric Research*, 2003].

[5] Prediction of clouds in mesoscale models used for air quality modeling applications has always been a difficult problem. Cloud processes on grid cell sizes of 4 km and greater are highly parameterized and uncertain. One of the weakest areas of meteorological models is the correct prediction of clouds at the correct location at the correct time. In air quality case studies, observations could conceivably be used to improve the specification of clouds. Unfortunately, standard weather service observations are not sufficiently dense to be used for cloud specification. However, geostationary satellite data can provide the desirable coverage with sufficient spatial resolution. The Geostationary Operational Environmental Satellite (GOES) has the capability to measure cloud properties such as optical reflectance down to scales of 1 km and cloud top heights to 4 km, and for timescales down to an hour or less. *Arola et al.* [2002] evaluated satellite retrievals of UV radiation over Europe and did not find any significant systematic bias in many of the methods used.

[6] In a previous study, *McNider et al.* [1998] used satellite-derived broadband cloud transmittance to correct NO_2 photolysis rates within the Regional Acid Deposition Model (RADM) [*Chang et al.*, 1987]. The case study for 3 August 1988 episode focused on the eastern United States. They concluded that the overestimation of the clouds by the meteorological model significantly reduced the photolysis rates as compared to the satellite-derived rates.

[7] In this paper, we present the results from incorporating satellite-derived transmissivity and cloud top height to provide the cloud properties needed in photolysis rate calculations, and use these revised photolysis fields in the CMAQ model. This is a first-order incorporation of the cloud effects. GOES visible and IR data collected and processed during the Texas Air Quality Study, 2000 (TexAQS2000) period are utilized. The impact of the satellite-based photolysis fields versus MM5-derived photolysis fields on ozone production is examined.

2. Methodology

[8] The method described in the following for cloud correction is based on the current formulation in CMAQ. As mentioned before, CMAQ uses a two step approach for photolysis rate calculations. First, clear sky photolysis rates are calculated, and then they are corrected for the cloud cover. While in the following the implementation within CMAQ is described, the method can be applied to any air quality model that uses a similar two step approach for the calculation of photolysis rates. In the following first a brief description of the current method used in CMAQ is presented. EPA's Models-3/CMAQ Science Document contains a detailed description of this approach [*EPA*, 1999]. Following this brief description, our approach is presented. The main issue explored here is the use of satellite-derived clouds as opposed to model-generated clouds for cloud correction. Therefore the technique presented here can be beneficial to any other model that uses model-generated clouds for cloud correction.

2.1. Current Method for Cloud Correction in CMAQ

[9] The method used for photolysis rate calculation and the subsequent cloud correction to those rates are described in EPA's Models3 Science document [*EPA*, 1999]. Photolysis rate (s^{-1}) is represented by:

$$J = \int_{\lambda_1}^{\lambda_2} \sigma(\lambda)\varphi(\lambda)F(\lambda)d\lambda \qquad (1)$$

where $\sigma(\lambda)$ (m^2/molecule) is the absorption cross section for the molecule undergoing photodissociation as a function of wavelength λ (μm); $\varphi(\lambda)$, the quantum yield (molecules/photon), is the probability that the molecule photodissociates in the direction of the pertinent reaction upon absorbing the radiation of wavelength λ; and $F(\lambda)$ is the actinic flux (photons/m^2/s/μm).

[10] By providing the actinic flux for clear sky, photolysis rates (J_{clear}) can be calculated by equation (1). In CMAQ, clear sky rates are then corrected for cloud cover. The cloud correction is based on *Chang et al.* [1987] and *Madronich* [1987] with some alterations as described in CMAQ Science Document [*EPA*, 1999]. A brief description of cloud correction as implemented in CMAQ is presented in the following. Below the cloud, the rate is corrected by:

$$J_{below} = J_{clear}[1 + f_c(1.6tr_c \cos(\theta) - 1)] \quad (2)$$

where f_c is the cloud fraction for a grid cell, tr_c is cloud transmissivity, and θ is the zenith angle. The above formulation leads to a lower value for the photolysis rates below the cloud, where the cloud transmissivity is reduced. Above the cloud, the photolysis rate is modified as:

$$J_{above} = J_{clear}[1 + f_c\alpha \cos(\theta)(1 - tr_c)] \quad (3)$$

Here α is a reaction-dependent coefficient that further modifies the rates above the cloud [*Chang et al.*, 1987]. This is to allow for the photolysis rate enhancement resulting from the reflected radiation from the cloud top. Within the cloud, the photolysis rates are obtained by interpolating between cloud base and cloud top values (which is a deviation from Chang et al.). Therefore, on the basis of the formulation above, the cloud transmittance and cloud fraction are required for calculating cloud correction for photolysis rates. Since in-cloud photolysis rates are interpolated, cloud base and cloud top heights must also be known.

[11] In CMAQ, the calculation of cloud transmissivity is highly parameterized [*EPA*, 1999]. The formulation is based on the parameterization suggested by *Stephens* [1978]. By obtaining cloud thickness (H_c) and liquid water content (w) the liquid water path (g/m^2) is calculated by:

$$LWP = wH_c \quad (4)$$

Then the broadband cloud optical depth (τ_c) as a function of liquid water path, assuming that the drop-size distribution within the cloud column is uniform, is calculated as [*Stephens*, 1978]:

$$\tau_c = 10^{.2633 + 1.7095 \ln[\log_{10}(LWP)]} \quad (5)$$

Finally, cloud transmissivity is calculated by:

$$tr_c = \frac{5 - e^{-\tau_c}}{4 + 3\tau_c(1 - \beta)} \quad (6)$$

where β is the scattering phase-function asymmetry factor [*EPA*, 1999]. In equation (6) it is further assumed that β is constant and has a value of 0.86. For optically thin clouds where $\tau_c < 5$ cloud correction is not performed. As evident from this formulation, even if the MM5-derived cloud fields were correct, there is some uncertainty in the calculation of cloud transmittance by equation (6) because of the assumptions used in different steps (as stated above).

[12] From GOES satellite observations, we are able to recover broadband cloud transmissivity and the cloud top height. Also, since GOES cloud mask algorithm can detect clouds (and the impact of subscale clouds) at 4 km resolution, an observed cloud fraction can be calculated for coarser grid cells as the fraction of cloudy pixels within a grid cell. Cloud base height is estimated as the local condensation level (LCL) from the temperature and mixing ratio profiles simulated by the mesoscale model. In this study, we replaced tr_c and f_c in equations (2) and (3) with the satellite-inferred quantities to perform the cloud correction.

2.2. Retrieval of GOES Broadband Visible Transmission and Cloud Top Heights

[13] The Infrared Measurement and Processing Group (hereinafter IR Group) at the National Space Science and Technology Center performed the satellite retrievals for this study. Currently, the IR group uses GOES Product Generation System (GPGS) to provide routine real-time retrievals of skin temperature, total precipitable water, cloud top pressure, cloud albedo, surface albedo and surface insolation for the use of meteorological and air quality models [*Haines et al.*, 2004]. As input, GPGS needs a first guess field for its retrievals and the model grid information if the product is to be used in a grid model. For this study, the MM5 simulation that was utilized for the CMAQ runs provided the required information to GPGS and the retrievals reflected the MM5 grid cell values.

[14] The algorithm used for the retrieval of albedo and surface insolation is the implementation of *Gautier et al.* [1980] method complemented by the improvements from *Diak and Gautier* [1983]. The method uses the information from GOES Imager visible channel (0.52–0.72 μm) at 1 km resolution and employs a clear and a cloudy atmosphere to explain the observed upwelling radiant energy. The model applies the effects of Rayleigh scattering, ozone absorption, water vapor absorption, cloud absorption, and cloud reflection. The effects of Rayleigh scattering are modeled after *Coulson* [1959] and *Allen* [1963] for the GOES visible band (radiant flux as viewed by the satellite) and for the bulk solar flux incident at the surface. Ozone absorption is modeled after *Lacis and Hansen* [1974]. Water vapor absorption is assumed to be negligible in both the surface and cloud albedo calculations, but accounted for when applying the total solar flux in the surface insolation calculation. Water vapor absorption coefficients are obtained from *Paltridge* [1973], and total column water vapor is assumed to be 25 mm and adjusted for solar zenith angle. Cloud absorption (for thick clouds) is assumed to be a constant 7% of the incident flux at the top of the cloud [*Diak and Gautier*, 1983].

[15] The surface albedo for the entire domain is calculated by using the clear-sky composite image. For the current study, a 20-day composite centered on the period of the case study was used. The single composite image records the minimum albedo value for each pixel for a given hour. Assuming that for any given hour during the day (for the entire month) each pixel experiences clear-sky at least once, the minimum value would represent the clear-sky value for that pixel. This formulation assumes that the visible channel surface albedo does not vary significantly within the time period of composite.

[16] The insolation is calculated as the sum of solar radiation incident at the surface from both direct and diffuse

sources and also includes the effect of attenuation by clouds. For the clear-sky case, the incident short-wave radiation at the surface is (1) the incident solar flux that is attenuated by Rayleigh scattering, ozone and water vapor absorption and (2) the surface reflected flux scattered back to the surface by Rayleigh scattering. With the surface albedo known, and the absorption and scattering processes estimated, the surface insolation is calculated directly.

[17] For the cloudy-sky, the satellite-derived radiant energy is the sum of atmospheric backscatter, reflection of the incident solar flux from the cloud surface, backscatter within the cloud by Rayleigh scattering, and the amount of surface reflection that reaches satellite after attenuation. Since the radiance at the satellite, the surface albedo, and estimates of the scattering and absorption are known, the radiation formulation can then be solved for the cloud albedo. In practice, the algorithm calculates a surface insolation using both the clear-sky and cloudy-sky formulations for a given scene. If the cloudy-sky calculation is greater than or equal to the clear-sky value, then the clear-sky value is used and the scene is assumed clear. This is consistent with the cloud albedo being near zero for clear-sky conditions. Since the effect of cloud albedo dominates in the insolation calculation, uncertainties in cloud thickness have been shown to produce only small effects on the surface insolation calculation [*Haines et al.*, 2004].

[18] Since the sum of cloud albedo (A_c), cloud absorption (a_c), and cloud transmittance is 1, then the broadband cloud transmittance is calculated as:

$$tr_c = 1. - (A_c + a_c) \tag{7}$$

The other needed vital information for our cloud correction is the cloud top height. A cloud top pressure is assigned to each cloudy pixel. GOES 11-μm window channel (of either the Imager or the Sounder) brightness temperature is used for this purpose. The clouds are assumed to be uniform in coverage and height over the GOES pixel. The brightness temperature for each cloudy pixel is referenced to the corresponding thermodynamic profile for the closest model grid. No attempt is made to correct the brightness temperature for the effect of water vapor above the cloud. The pressure assignment is similar to that used by *Fritz and Winston* [1962] and applied by *Jedlovec et al.* [2000]. Log linear interpolation is used between model vertical pressure levels to assign a corresponding pressure for the cloud top temperature.

[19] The approach works well for opaque clouds where the cloud emissivity is close to unity and emission (measured by the satellite) comes primarily from the cloud top. Typical pressure assignment errors are on the order of 25–50 mbar (0.5–2.0 K). For nonopaque clouds such as thin cirrus, emission from below the clouds is detected by the satellite and cannot be separated from cloud emission without knowledge of the cloud emissivity. The bias would be greatest for low clouds.

[20] For air quality applications, however, since the focus is on the boundary layer, the error in the cloud top pressure for the opaque clouds does not pose a significant problem. Furthermore, in our technique the cloud top height is only used for determination of the atmospheric layer in which photolysis rates are being interpolated, and does not impact the correction made to the photolysis rates within the boundary layer (as the transmittance is estimated directly from the satellite observations). In addition, the determination of cloud top in the model is limited by the vertical resolution of the model, which usually is too coarse in the free troposphere. For the nonopaque clouds, the cloud transmissivity is large and therefore the modifications to photolysis rates are small and thus the impact of the error in the cloud top height is further reduced. Figures 1a and 2a illustrate a situation on 24 August 2000, where the satellite observation indicates most of the domain is cloudy, yet in fact only the cloud mass over the Galveston Bay area is opaque. For most of the domain, the clouds are almost transparent and the retrieved cloud transmittance is close to 1. For low transparent clouds with unrealistic cloud top pressure, we allow for a thin cloud above the cloud base (only one model layer thick).

2.3. Implementation Within CMAQ

[21] The current setup for CMAQ calculates the clear sky photolysis rates in a preprocessor and provides a tabular input to the Chemical Transport Model (CTM) [*EPA*, 1999]. The meteorological data, including cloud information derived from MM5 predictions, is prepared in a Meteorology-Chemistry Interface Processor (MCIP) for the use in CTM. Within the CTM the attenuation to clear sky photolysis rates due to the presence of clouds is performed on the basis of the input information from the meteorological model. We have made modifications to the MCIP to replace the MM5-derived (hereinafter referred to as MCIP clouds) cloud information with the satellite observations.

[22] In the presence of satellite observations, cloud fraction in MCIP is replaced with the observed cloud fraction. From cloud top temperature (or pressure as discussed above), the corresponding CTM layer is identified as the cloud top layer. Model surface temperature and mixing ratio are used to calculate the lifting condensation level, and is used as the cloud base height. Within the CTM, when satellite-retrieved transmissivity is present, the standard parameterization is bypassed and the satellite observations are used directly in equations (2) and (3).

3. Model Simulations

[23] We implemented the technique described above in the CMAQ modeling system to perform a set of simulations for 12 and 4 km resolution domains over Texas for the period of 24–31 August 2000. The 12 km domain covers the eastern half of Texas, Louisiana, Mississippi, southern part of Oklahoma and Arkansas, and the southwestern corner of Tennessee. The first set of simulations utilizes CMAQ in its standard configuration, and is used as the control case (hereinafter referred to as CMAQ_base) for comparison. The second set of simulations (hereinafter referred to as CMAQ_sat) uses the satellite-derived cloud information. Both sets of simulations use the same meteorological information from a single MM5 run.

[24] The control MM5 simulation was configured to use FDDA gridded nudging, Dudhia moisture scheme, Grell convective parameterization, Medium Range Forecast (MRF) PBL scheme, RRTM radiation scheme, shallow convection scheme, and five-layer soil model. Grell cumu-

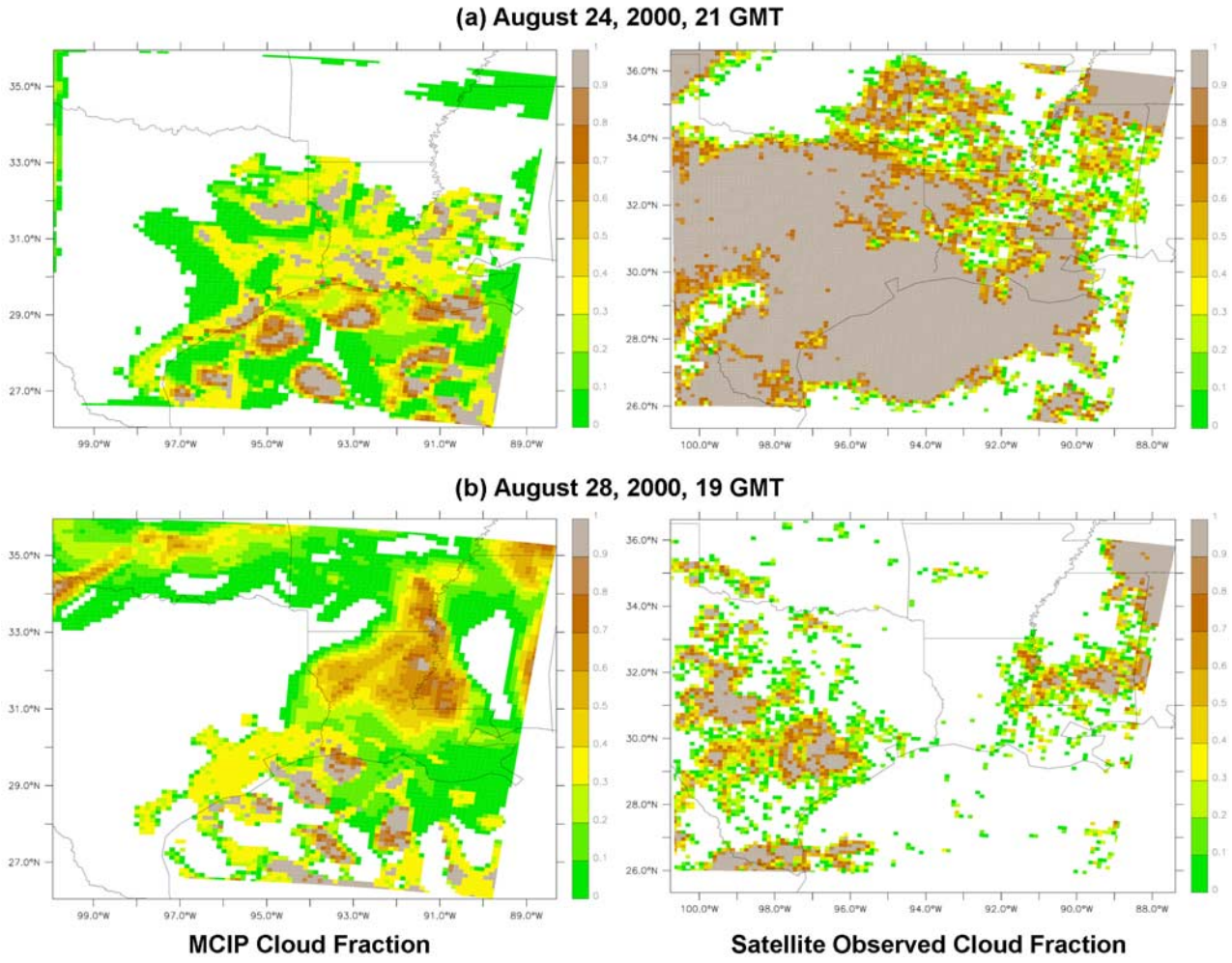

Figure 1. MM5 predicted and satellite observed cloud fields for (a) 24 August 2000, 2100 UT, and (b) 28 August, 1900 UT.

lus parameterization has proven to be useful for smaller grid sizes (10–30 km). It tends to allow a balance between resolved scale rainfall and convective rainfall [*Grell et al.*, 1991; *Grell*, 1993].

[25] CMAQ (version 4.3) was configured to use piecewise parabolic method for advection, multiscale horizontal diffusion and eddy vertical diffusion, 3rd generation aerosol model and 2nd generation aerosol deposition model, RADM cloud model, and SMVGEAR chemical solver. Carbon bond IV (CB4) chemical mechanism [*Gery et al.*, 1989], including aerosol and aqueous chemistry is utilized to describe atmospheric reactions. The model uses 21 layers, with about 10 layers within the daytime boundary layer. The emissions for this study are based on EPA's 1999 National Emissions Inventory (NEI99, version 2).

4. Results and Discussion

[26] As described in the previous section, the meteorological information to drive CMAQ was obtained from a single MM5 run. This means that there is no change in the dynamic fields for the CMAQ simulations and the differences between CMAQ_base and CMAQ_sat simulations are only due to the impact of observed clouds on the photochemistry. This inconsistency also impacts the heterogeneous processes in the model. In the areas where the model is underpredicting clouds, use of observed clouds reduces the errors in the gas phase chemistry but the accompanying heterogeneous chemistry in the cloud layer is nonexistence in the model. On the other hand, when the model overpredicts clouds, our technique will increase photolysis rates throughout the atmospheric column while the heterogeneous processes in the model are still active. Such errors in the current study are unavoidable (as they are inherent from the control MM5 simulation) and can only be corrected if the model is dynamically consistent with the observations. The current study is only focusing on the radiation impact of clouds on the photochemistry and the impact of cloud dynamics will be pursued in the subsequent papers.

[27] It should be noted, however, that the uncertainty due to the impact of cloud dynamics on the vertical transport of the pollutants is also important and needs to be investigated. For example, on the afternoon of 24 August 2000, convective clouds developed over the Galveston Bay and expanded toward north/northwest. This feature was absent in the MCIP cloud fields, meaning that the vertical transport of pollutants over the Bay area into these convective cells is

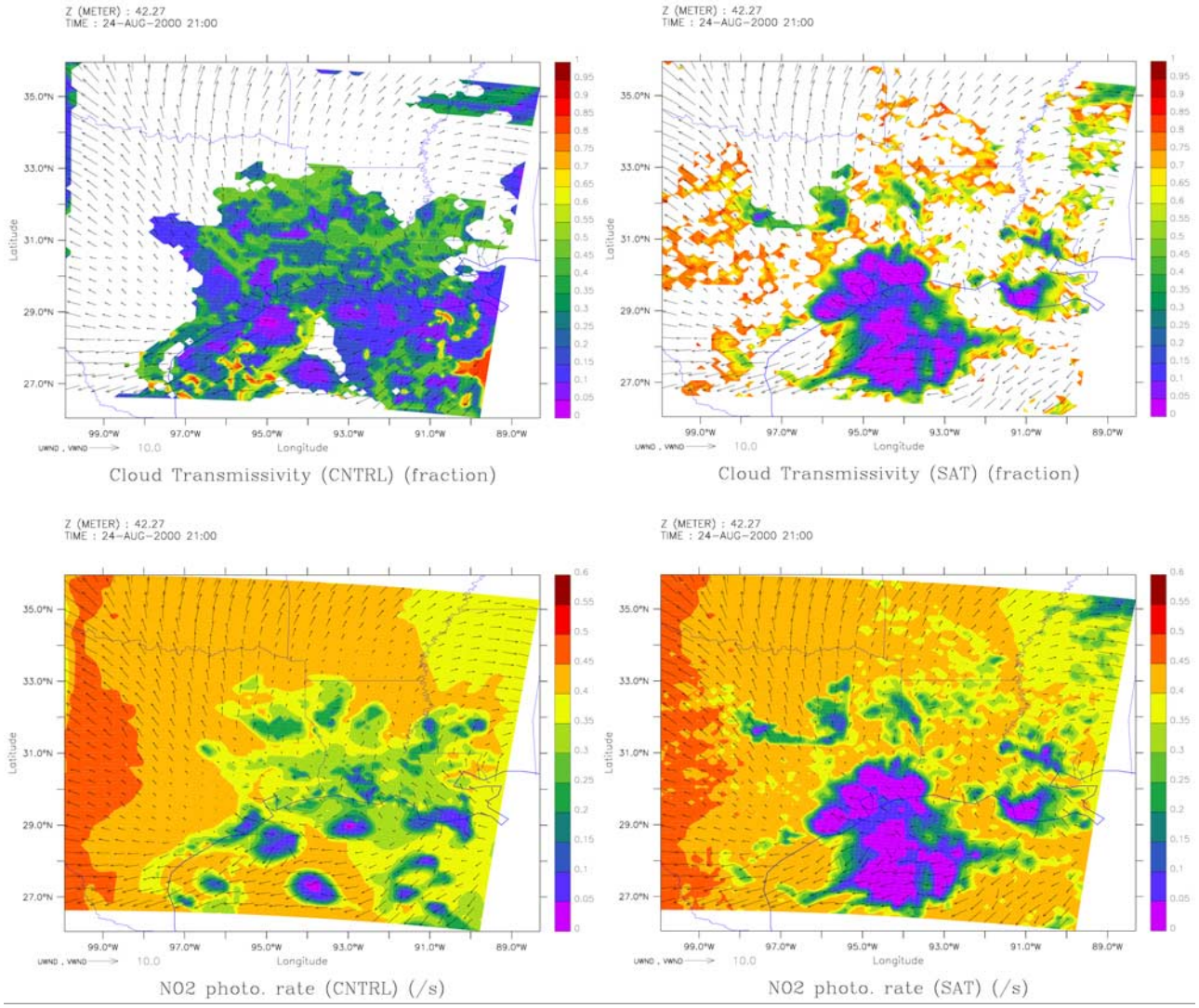

Figure 2. Cloud transmissivity and corresponding NO_2 photolysis rates for 24 August 2000 at 2100 UT from CMAQ_base and CMAQ_sat simulations at the surface (first model layer).

missing in our simulations. While our method corrects for the impact of the observed convective clouds on the photochemistry, there are still errors arising from the lack of accurate vertical distribution of pollutants due to errors in the dynamics. Therefore here we only emphasize on model-to-model comparisons to illustrate the first-order photochemical impact of including the observed clouds. In the second part, however, we present comparisons with selected observations to illustrate that the large differences seen in the model-to-model comparison are indeed real and our technique is greatly improving the model performance.

4.1. Model-to-Model Comparisons

[28] Texas and surrounding areas were extremely dry for the period of this study, and perhaps not the best case to show the benefits of utilizing GOES information. Nevertheless, there was sufficient cloudiness to illustrate the impact of observed clouds on the photochemical model predictions. Figure 1 displays two different cases in which the disagreement between MCIP cloud fields and GOES observations are depicted. In the case of 24 August 2000, MCIP indicated clouds in the south/southeastern part of the domain with most of it being subgrid scale (with cloud fractions less than 1) with only a few small areas of grid scale clouds over land. In contrast, satellite observations indicated large area of cloudiness extending from south/southeast to the northwest part of the domain. Satellite observation also indicated clouds in the northeast and northern parts of the domain that were absent in the MCIP fields. However, as indicated in Figure 2, the broadband transmissivity for most of the observed clouds for this day is high, meaning that most of the clouds are not opaque and should not affect the photolysis rates significantly. However, the area around Galveston Bay, including Houston, is covered with thick clouds that are missing in the MCIP fields. This is significant, as this area is the major source of emissions for ozone precursors.

[29] An error in the prediction of opaque clouds over the emission sources has major consequences. Opaque clouds (as seen in Figure 2) can significantly alter the cloud transmissivity and, thus, the photolysis rates. Over the source regions, an alteration (reduction in this case) in

Figure 3. An image of Houston-Galveston Bay area. Locations at (29.7, −95.3) (labeled A) and (30, −95.6) (labeled B) are marked with red circles.

the photolysis rate has both a direct and an indirect impact on ozone chemistry. First, by slowing down the photochemistry, lower photolysis rates inhibit ozone production in the immediate vicinity of emission sources (direct impact). Second, because of the suppression of photochemistry, lifetime of ozone precursors is increased and the precursors can be transported to the regions where the air mass has a different chemical composition (indirect impact). The indirect impact can take many forms depending on the type of the cloud and the time of occurrence. These include the impact on the boundary layer air further downwind (for the clouds with weak vertical motion during the day), the accumulation of the precursors in the residual layer (clouds late in the day), or alteration in the chemical composition of free troposphere (convective cells with strong vertical motion).

[30] In the case of Galveston Bay region, nitrogen oxides ($NO_x = NO + NO_2$) and volatile organic compounds (VOC) are coemitted (on the regional scale). Therefore, in this region under clear skies, ozone is rapidly produced while NO_x is transformed to products such as nitric acid (HNO_3) and peroxyacetyl nitrate (PAN). The inhibition of the photochemistry in the presence of clouds on the other hand directly impacts the rapid formation of ozone in this area and by doing so both NO_x and VOCs remain active for a longer period of time. In short, such an event alters the chemical aging of the air mass, and the air mass continues to have the potential of producing ozone for a longer period of time during transport.

[31] Another indirect impact of the clouds in this area is the alteration in partitioning of nitrogen oxides and the impact on nitrogen budget due to surface removal. This is caused by the disparity between the deposition velocity of NO_x and the nitrates that are produced from oxidation of NO_x. Under clear skies, as indicated before, NO_x in this region undergoes a chemical transformation and produces nitrates such as HNO_3 and PAN. In the presence of thick clouds, because of the reduction in the photochemical activities, nitrogen monoxide (NO) rapidly consumes ozone (O_3) and produces nitrogen dioxide (NO_2) while the production of HNO_3 and loss of NO_x due to chemical transformation is reduced. In this case while the partitioning of NO_x between NO and NO_2 has been altered, there is a net increase in NO_x due to its direct emissions.

[32] Therefore, in one case, under clear conditions over the Galveston Bay area in the control case, more O_3, HNO_3, PAN and other nitrates are produced in the expense of NO_x. However, under cloudy conditions (satellite assimilation case), because of the slowing down of the photochemistry, most of the NO_x will remain intact and will not be lost in the ozone production to produce nitrates. The rate of surface removal for NO_x is an order of magnitude less than that of nitric acid [*Biazar*, 1995]. Therefore, in control simulation there is a much larger loss of total reactive nitrogen ($NO_y = NO_x + HNO_3 + PAN +$ other compounds produced from the oxidation of NO_x) than the assimilation simulation.

[33] To show such an indirect impact, a grid point close to the bay (southeast of Houston at 29.7°N, 95.3°W, marked A on the map in Figure 3) was examined. Comparing the accumulated hourly surface deposition from the two simulations (CMAQ_base versus CMAQ_sat) for NO_x and HNO_3 reveals that the absence of clouds (in the control case) increased the surface removal of HNO_3 for several hours for up to 9 g/hectare/hr (Figure 4). The loss of nitric acid positively correlates with the increased ozone production and increased NO_2 photolysis rate at this location (as shown in Figure 5a) and is a result of the increased HNO_3 production due to active photochemistry. The inclusion of clouds resulted in less than 1 g/hectare loss of NO_x in this case.

[34] In contrast to the 24 August case, on 28 August, MCIP indicates a large area of cloudiness over western Mississippi, southern Arkansas, and Louisiana extending to the south Texas (Figure 1b). This is absent in the GOES observations. GOES observations indicate subgrid cloudiness in the western part of Texas. From Figure 2b it can be seen that these clouds are highly transparent and do not alter the photolysis rates significantly. Therefore, in this case we have a significant ozone formation in the vicinity of the emission sources that would be absent in the control simulation.

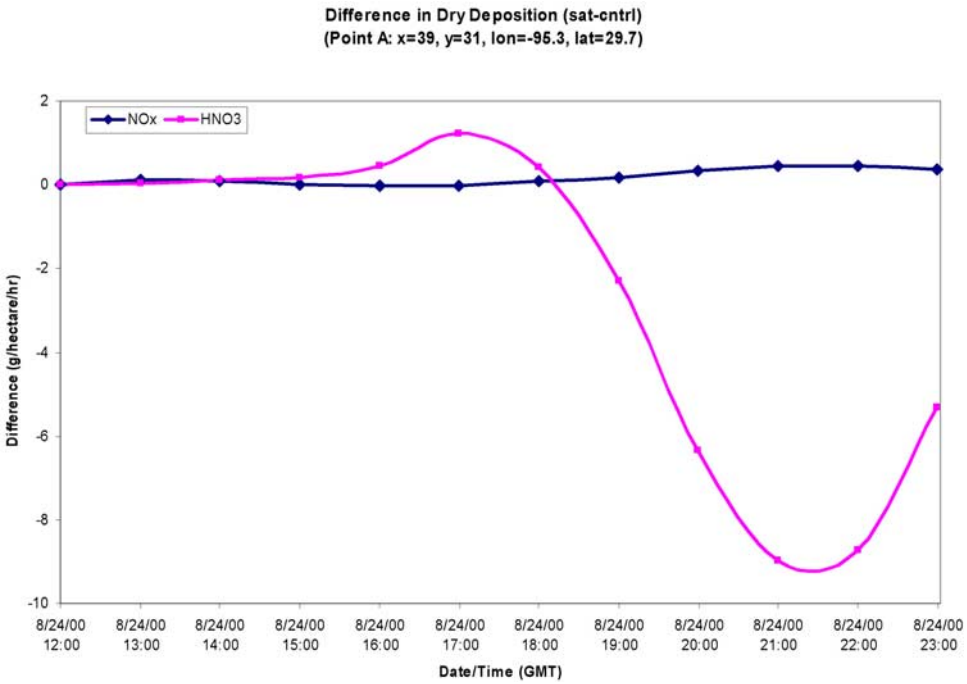

Figure 4. Hourly differences in NO_x and HNO_3 surface removal from the two simulations (control versus satellite assimilation) for point A over Houston-Galveston Bay area for 24 August 2000.

[35] The impact of such alterations in the photolysis rates on the local atmospheric chemical composition can be substantial, especially on the chemical species with the shorter photochemical lifetime. Figures 6 and 7 exhibit the largest differences in NO, NO_2, NO_x, and ozone between the assimilation and control simulations over the entire period of study for the 12 km domain. Figures 6 and 7 represent the extreme cases of discrepancy between control and assimilation simulations, and these extremes may not occur at the same time. However, examining the time series of larger values indicated that they occur about the same time and represent a shift in NO_x partitioning. This is evident in spatial patterns in Figure 6 as the negative/positive values for NO are colocated with the positive/negative values of NO_2. The areas marked with a large negative NO difference between the assimilation and control correspond to the situation where MCIP indicates overprediction of clouds and therefore most of NO is converted to NO_2 (and vise versa). These areas are confined to the large source regions, as evident for example over the Houston-Galveston Bay area, indicating a much faster photochemical activity and rapid ozone formation.

[36] For the NO_2 case (Figure 6b) there are broader areas of large discrepancy. Over the Texas region, this indicates the transport of NO_x outside the source region where the lifetime of NO_2 is increased. This is perhaps due to the transport and dilution of the air mass outside the source region and mixing with an air mass of lower VOC where the rapid ozone formation is inhibited. The evidence for the above statement can be seen in Figure 7b in which largest ozone differences are depicted. Also, the large discrepancy in NO_x (Figure 7a) to the north of Houston is indicative of NO_x transport out of the source area due to inhibition of photochemistry in the presence of clouds.

[37] Figure 7b also indicates that there are times that the impact of our method on ozone concentration can be quite high (as much as 60 ppb). While these extreme cases are mostly localized in space and time, sustained differences of several ppb over broader areas are more common. Comparing the extreme values of NO_x and ozone, there is a good correlation between higher ozone concentrations in the assimilation run and lower NO_x concentrations (and lower NO_2 concentrations). This indicates the presence of observed clear sky in contrast to MCIP indicating overpredictions of clouds. Therefore the assimilation run produces more ozone and nitrates at the expense of NO_x. On the other hand, underpredictions of clouds in MCIP cloud fields resulted in higher ozone values in the control run for the east/southeast and northern part of Louisiana and a large part of central Texas.

[38] As indicated in Figures 6 and 7, the impact of alterations in the photolysis rates on the local atmospheric chemical composition can be substantial, especially on the chemical species with the shorter photochemical lifetime. It should be noted, however, that the domain-averaged differences only show a maximum of 2 ppb for 26 August and are mostly between ±1 ppb for other days. Domain-averaged differences also exhibit a diurnal variation with higher predicted ozone for the assimilation run. This indicates that in this case study the overall impact of clouds in the two simulations over the 12 km domain is not drastically different, meaning that we have had as much underpredictions as we had overpredictions. This is an indirect way of comparing the impact of total cloud cover in the two simulations and concluding that they are not very different. However, the large differences in ozone concentration in

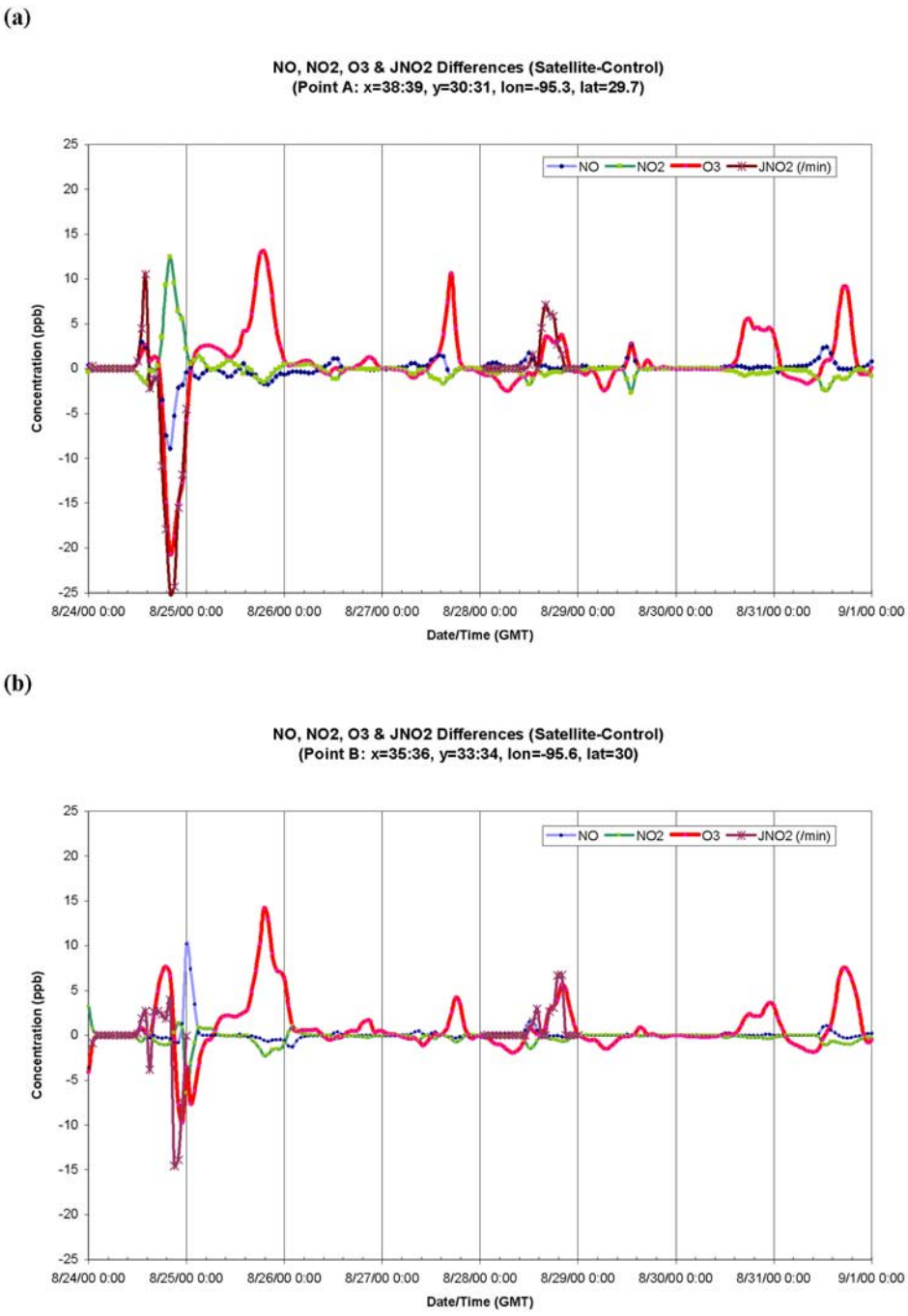

Figure 5. Differences between NO, NO_2, O_3 (ppb) and JNO_2 (/min, only for 24 and 28 August) between satellite cloud assimilation and control simulations for (a) grid cell A and (b) grid cell B (as marked in Figure 3) over Houston-Galveston area.

Figure 7, for example, indicate that the two simulations are very different in the temporal and spatial distribution of the clouds.

4.2. Houston-Galveston Bay Area and the Case of 24 August

[39] As evident from Figure 6, there are large differences between the two simulations over Houston-Galveston Bay area. In particular, there seems to be a sharp contrast between the air to the southeast of Houston and that of north/northwest of Houston. We picked two representative grid cells for these areas to be examined in more detail. The cells are marked with red circles in Figure 3. The coordinate for the cell to the southeast of Houston is 29.7°N, 95.3°W (marked A), and the cell to the northwest has a coordinate of 30°N, 95.6°W (marked B).

[40] Figure 5 illustrates the differences in NO, NO_2, and O_3 between the two simulations for these two grid cells.

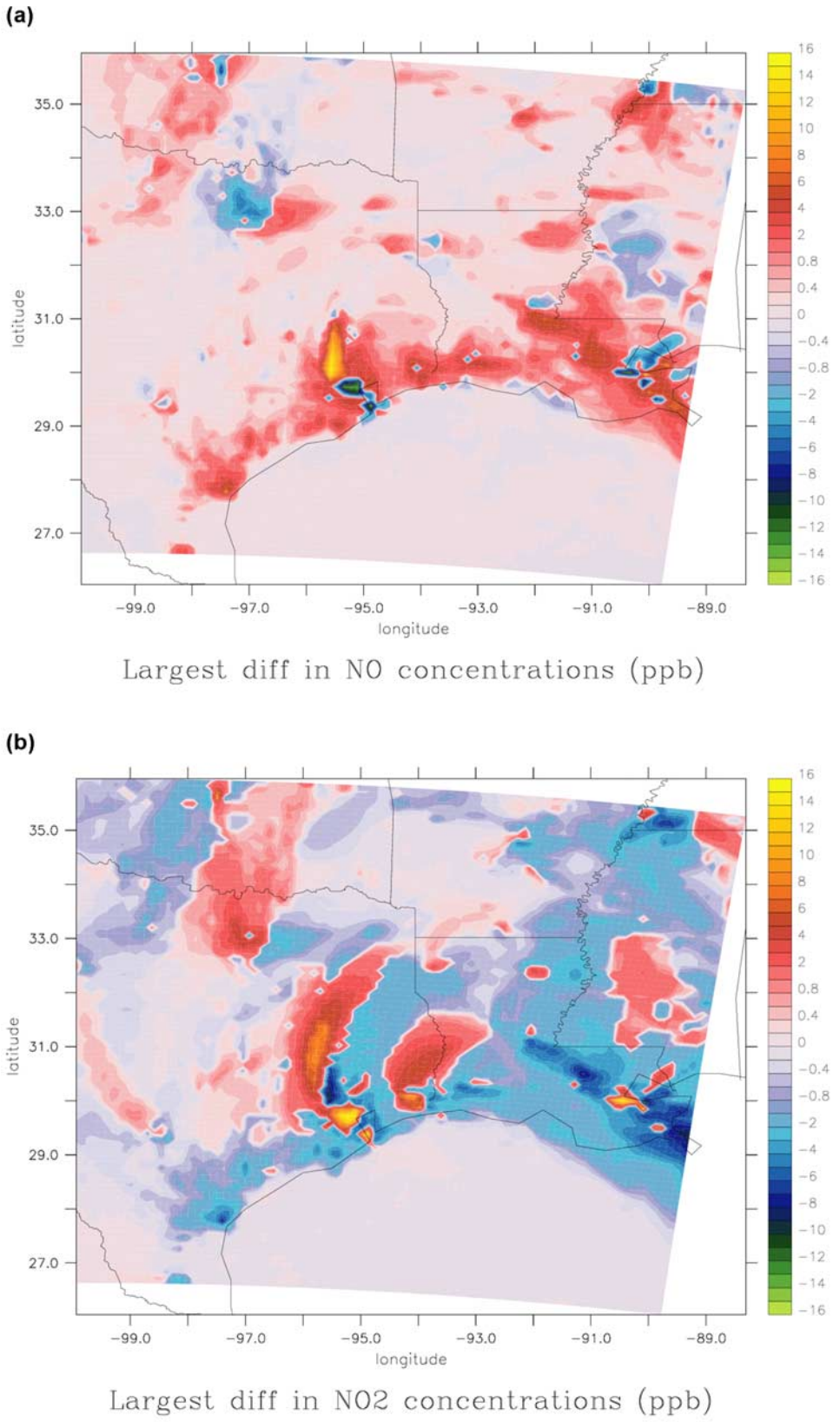

Figure 6. Largest differences in (a) NO and (b) NO_2 between assimilation and control simulations (assim-control) for the entire period of study covering from 0000 UT, 24 August 2000, to 0000 UT, 1 September 2000.

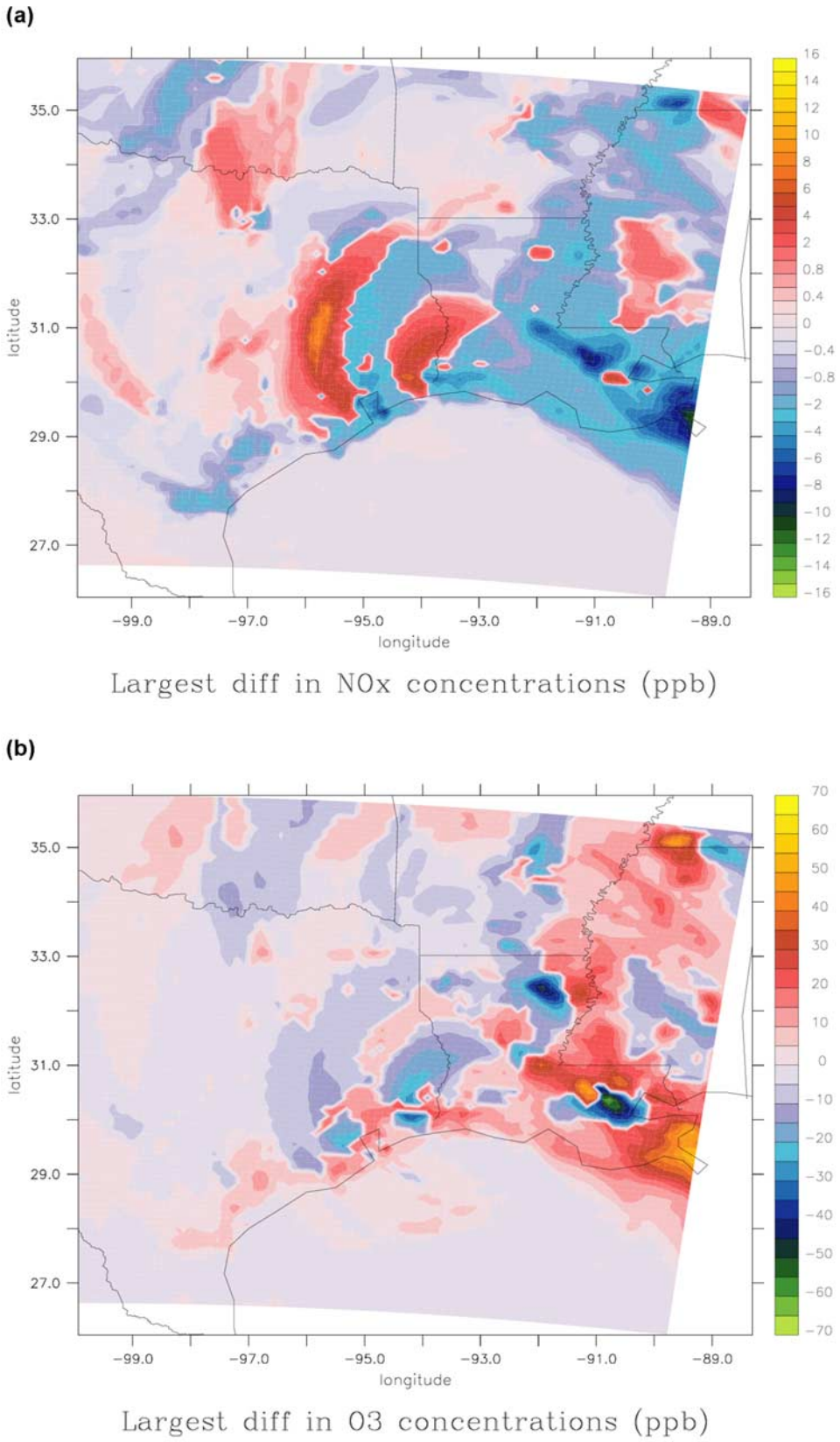

Figure 7. Largest differences in (a) NO_x and (b) O_3 between assimilation and control simulations (assim-control) for the entire period of study covering from 0000 UT, 24 August 2000, to 0000 UT, 1 September 2000.

Figure 5 also shows the differences in NO_2 photolysis rates between the two simulations for 24 and 28 August. Since the photolysis rates are not one of the standard outputs from the model, they were not saved for the entire period of simulation. However, the available data for both days clearly emphasizes the direct impact of the clouds.

[41] Interestingly, the extreme differences noted in Figure 6 for these cells appear to be from 24 August. This also coincides with the extreme difference in O_3 (Figure 7b) for point B. While the differences on 24 August are extreme, large differences are observed on many days for both grid cells. Almost in all cases a good negative correlation exists between O_3 differences and that of NO_2, indicating that most of these daytime differences are due to a discrepancy between modeled and observed clouds, and are the result of alterations in photochemical activity. The difference in O_3 is more pronounced than that of NO and NO_2, since in the control run not only O_3 production has been abated, but also at the same time O_3 is being consumed by NO to produce NO_2. In some cases, as in the case of 29 August, most of the difference seen is due to O_3 consumption by NO and the additional photochemical production is negligible. It should be noted, however, that our emissions over Houston-Galveston area could be low with respect to anthropogenic hydrocarbon emissions [*Allen et al.*, 2002]. If this proves to be the case, an increase in the hydrocarbon emissions would result in even higher discrepancy between control and assimilation simulations. The increase in the hydrocarbon emissions would expedite the photochemical activity with respect to ozone formation in the cloud-free areas.

[42] On 24 August, in midmorning to early afternoon period, model dynamics indicate a nicely formed see breeze that extends deep inland. The flow generally has a curvature, starting as an easterly/southeasterly flow offshore and turning to a southerly flow over land. Later in the afternoon, the inland flow becomes westerly and a convergence zone forms along the coast. In particular, over west/southwest of Houston-Galveston (HG) area the winds are calm after 2000 UT. About this time, satellite observations indicate the formation of the convective cells from south/southeast of HG area which later advances inland toward north/northwest. This created a situation in which the emissions to the southeast of HG were accumulating in the model as the cloud correction (according to the satellite observations) took place. The extreme values for point A occur at 2100 UT.

[43] In the control simulation, only about 5 ppb of ozone is produced (net change due to all the processes) from 1800 to 2100 UT (going from 18 ppb to 23 ppb). In the presence of clouds in the assimilation simulation, most of the ozone is consumed by NO producing NO_2 and creating large differences seen in Figure 5. As mentioned earlier, since the surface removal of NO_x is slower than that of HNO_3, most of the NO_2 in this air mass (50 ppb for the grid cell A) remains intact and will be converted instantly back to O_3 as soon as it is exposed to sunlight.

4.3. Verification of Model Results

[44] Up to this point we have compared the results from the satellite assimilation simulation against the control simulation, in which CMAQ in its standard configuration was applied. Now, the question is that while the differences in concentrations of ozone and nitrogen oxides between the two simulations are large, are these differences real and have we been able to correct model errors of the same magnitude? In other words, can we verify these results against observations and show that model predictions have improved?

[45] We acknowledge that the emissions used in this study need improvement and the uncertainties arising from the problems with the emissions are high. Nevertheless, for an area impacted by the cloud cover (or lack of it), we expect to see a variation in the concentrations that is more in line with the observations. Our hypothesis was that, for the areas impacted, the errors due to incorrect cloud cover in the model far exceed the errors caused by inaccurate emissions. To test this hypothesis, we compared ozone concentrations from the two simulations (CMAQ_base versus CMAQ_sat) with EPA's AQS (Air Quality System, http://www.epa.gov/ttn/airs/airsaqs) observations for the entire period of simulations.

[46] The overall large-scale spatial distribution of the predicted ozone for both the control and satellite assimilation simulations generally agreed with observations. Figure 8 depicts a snapshot of CMAQ_sat predictions at 2100 UT (1600 LT) on 24 August 2000. The model is able to predict the low ozone concentrations next to the Houston-Galveston Bay as well as the high ozone concentrations in the Dallas area. However, model predictions of high ozone concentrations in many rural areas and smaller towns cannot be substantiated because of the large gaps in the observational network.

[47] By using satellite clouds, the bias (mean error) for surface ozone predictions was reduced by 26%, from −4.05 to −2.99, while the RMSE was reduced by 3%. The predictions of peak ozone were improved by 1%. The domain-average predictions of peak ozone exhibit an insignificant improvement, but examination of the individual sites impacted by cloud misalignment indicate a much greater improvement. While these statistics indicate an improvement in ozone predictions, they are unable to show the full impact of satellite assimilation. Several factors affect large-scale statistical evaluation for this study. First, there are large data voids in the observational network, and since there are large spatial variations in surface ozone, performing objective analysis to fill in the gaps carries significant uncertainties. Second, most of the monitors are located in the vicinity of urban centers where they are largely impacted by local emissions and local weather. Therefore several monitors that may reside within one model grid can exhibit large variations (up to 50 ppb for ozone). In such cases doing a simple averaging for the cluster of observations will not suffice, especially since the model also indicates a large spatial gradient from one cell to another (urban to rural). Additionally, the problems with emissions, lateral boundary conditions for a relatively small domain, and lack of clouds for a significant part of this study also contributed to errors over the entire domain. Such errors will lead to modest statistics that conceal the improvements at individual sites impacted by observed clouds. Therefore, to test our hypothesis we evaluated model predictions over selected locations where the cloud impact was significant.

[48] For the selection of locations we referred to Figure 7b and identified the areas where the differences between the

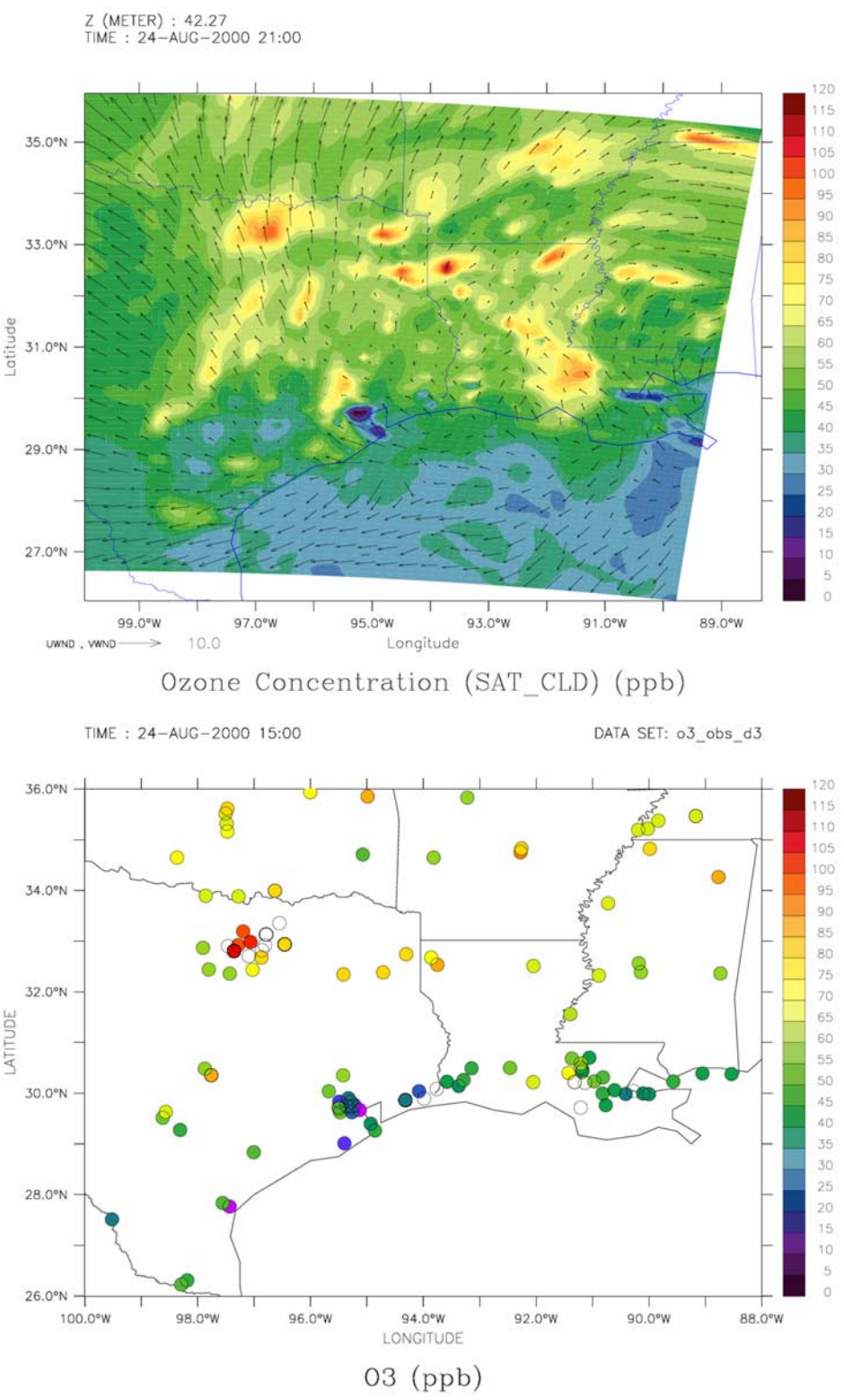

Figure 8. Model predictions of ozone versus EPA's AIRS observations for the 12 km domain on 24 August 2000, 2100 UT.

two simulations were the largest. In those areas, we picked the grid boxes that contained an observation site. Some of the largest differences occurred in the eastern part of the domain and over southern Mississippi and southeastern Louisiana. We could identify three grid boxes in that region fulfilling our requirements, namely two locations over New Orleans area and one over south Mississippi. As evident from Figure 7b, some of the extreme underpredictions and

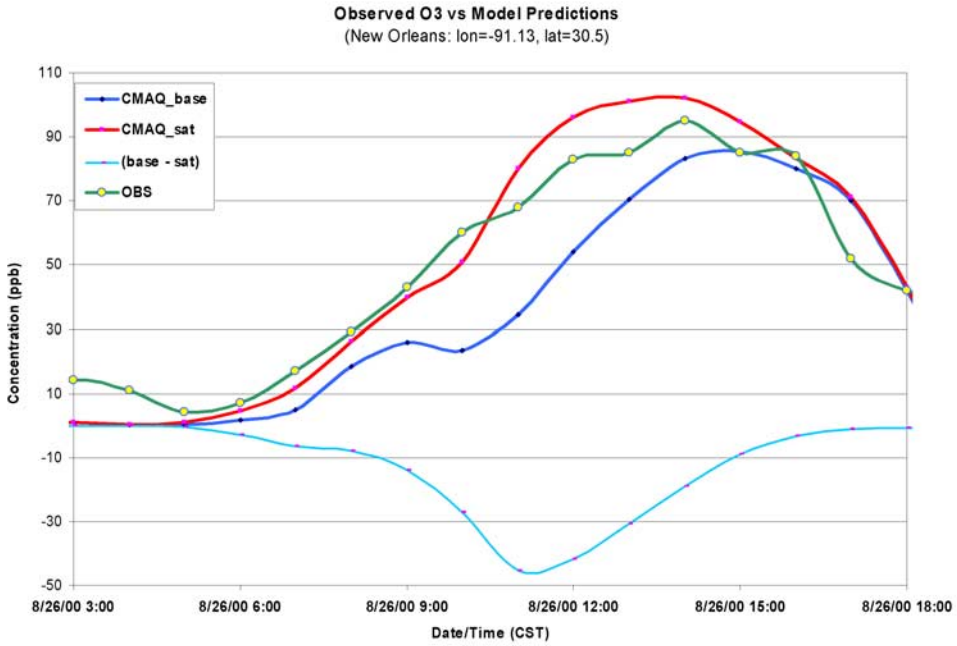

Figure 9. Time series of ozone predictions versus observations (OBS) for a location near New Orleans on 26 August 2000. CMAQ_base is the control simulation, and CMAQ_sat is the satellite assimilation simulation. The light blue line shows the difference between the two model simulations. The control simulation underpredicted ozone by about 35 ppb.

overpredictions of ozone by the base simulation occurs over this region. While these extremes do not occur at the same time, having the largest underpredictions and overpredictions of ozone in the same area indicates the importance of cloud effects in the source regions.

[49] Figure 9 shows the time series of ozone concentrations from the two CMAQ simulations plotted alongside observations on 26 August 2000, for a location over the New Orleans area. Just before the sunrise, both the model simulations drop to values close to the observation. However, after the sunrise, ozone concentration in the control run does not increase at the same rate as the observation. The slow rate of the increase and the subsequent decrease in the ozone concentration is due to the overprediction of clouds in MM5-derived fields for this location. Since this location is impacted by high NO_x concentration, a reduction in the photochemical activity due to the overcast sky causes ozone consumption and therefore a reduction in ozone concentration. On the other hand, the satellite assimilation simulation (CMAQ_sat) for this location on 26 August indicates a better agreement with the observations. Clearly in this case the underprediction of up to 35 ppb in ozone concentration is due to the MCIP indicating overprediction of clouds and the use of satellite observed clouds has been able to correct this error.

[50] The second example, as depicted in Figure 10, shows a scenario in which CMAQ_base is overpredicting ozone concentrations because of the lack of clouds in MCIP fields while in fact the sky is cloudy. In this case both the control and assimilation simulations overpredict ozone concentrations for most of the day over a location near New Orleans on 31 August. This is perhaps due to the errors in the emissions for this location. However, in the afternoon, as the clouds move over this location, CMAQ_sat exhibits a sharp decrease in ozone concentration similar to that of the observation while the concentrations in the control run remain high. At 1700 LT, ozone concentration from the assimilation run agrees with the observation while the control simulation overpredicts ozone by 58 ppb. This also speaks to the impact of cloud correction and indicates that a reduction in photochemical activities in this location is enough to correct the large model overprediction of ozone.

[51] The third case is from a location in southern Mississippi on 31 August. As depicted in Figure 11, this case indicates underpredictions of ozone by the CMAQ_base run while the CMAQ_sat run again shows a better agreement with the observations. For this location the observations exhibit some variations which could be due to passing plumes that are not captured well in the simulations. However, the largest discrepancy, which is an underprediction of about 35 ppb at 1600 LT in the control run, is due to the overprediction of clouds in MM5-derived fields. Again in this case we observe a reasonable agreement between CMAQ_sat and observations at that time.

[52] For all these locations during the nights CMAQ_base and CMAQ_sat are generally in agreement and their deviation from measured concentrations (that are due to other errors in the model) is smaller than the errors introduced because of incorrect cloud cover specification. Indeed in most of the domain, when there was a discrepancy between MCIP cloud fields and that of the observations, the largest errors could be attributed to the impact of clouds.

5. Conclusion

[53] In this study, satellite-retrieved cloud transmissivity, cloud top height, and observed cloud fraction were used to

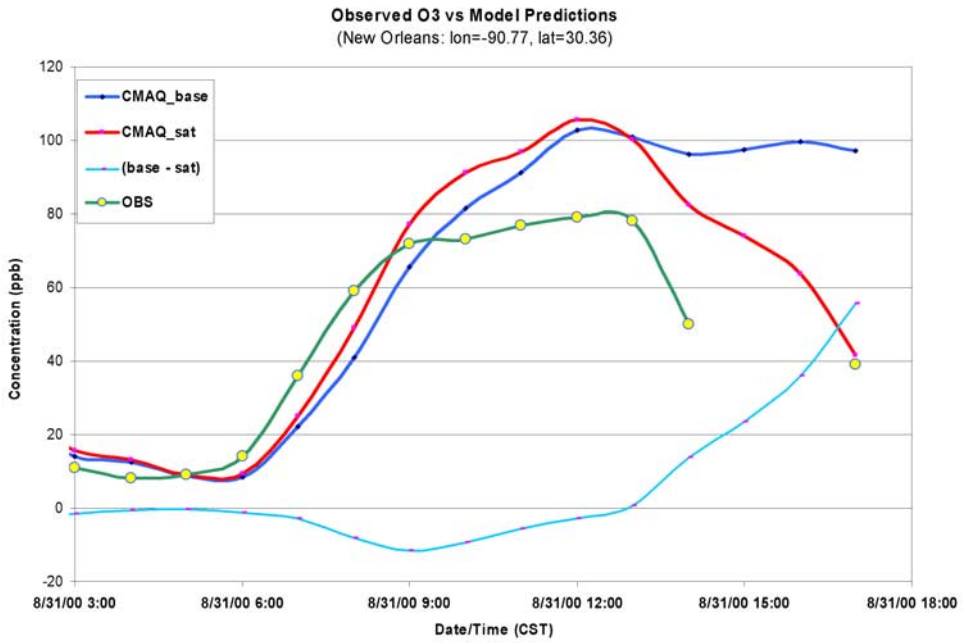

Figure 10. Time series of ozone predictions versus observations (OBS) for a location near New Orleans on 31 August 2000. CMAQ_base is the control simulation, and CMAQ_sat is the satellite assimilation simulation. The light blue line shows the difference between the two model simulations. Control simulation overpredicted ozone by about 58 ppb.

correct photolysis rates for cloud cover in CMAQ. The results from CMAQ simulations using this method were compared with simulations that used standard MM5-derived cloud fields as input. The simulations were performed with 4 and 12 km grid cell sizes over Texas, extending east to Mississippi, for the period of 24 to 31 August 2000.

[54] The results reveal that lack of observed clouds in the model can drastically alter the predicted atmospheric chem-

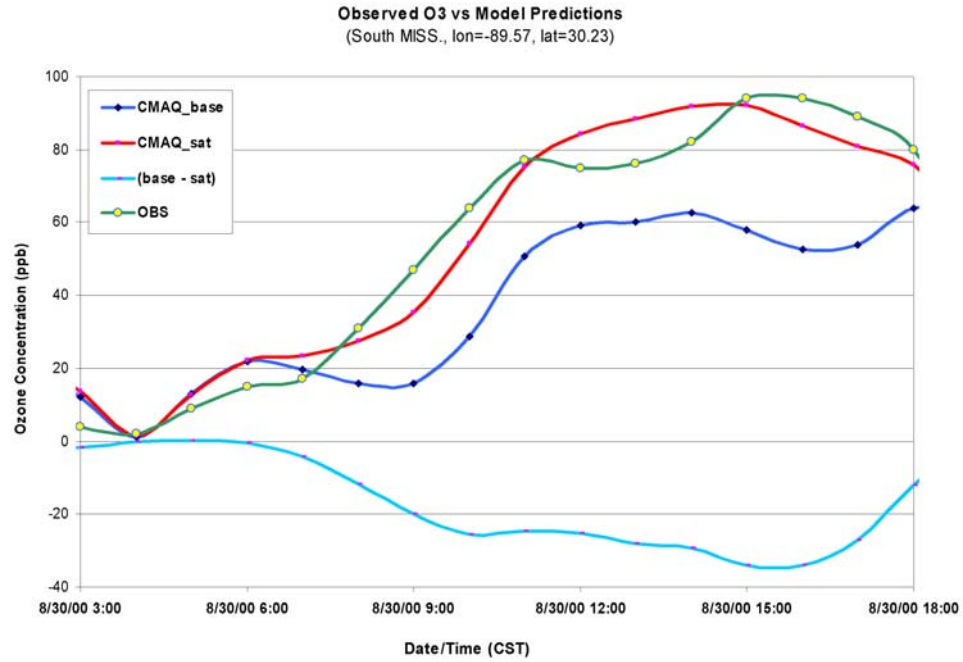

Figure 11. Time series of ozone predictions versus observations (OBS) for a location in south Mississippi on 30 August 2000. CMAQ_base is the control simulation, and CMAQ_sat is the satellite assimilation simulation. The light blue line shows the difference between the two model simulations. Control simulation underpredicted ozone by about 35 ppb.

ical composition within the boundary layer and exaggerate or underpredict ozone concentrations. Cloud impact is acute and more pronounced over the emission source regions and can lead to large errors in the model predictions of ozone and its by-products. Clouds also increased the lifetime of ozone precursors leading to their transport out of the source regions and causing ozone production farther downwind. Longer lifetime for NO_x and its transport out of the source regions and over regions high in biogenic hydrocarbon emissions (in the eastern part of the domain) led to increased ozone production that was missing in the control simulation. Over Houston-Galveston Bay area, the presence of clouds altered the chemical composition of the atmosphere and reduced the net surface removal of reactive nitrogen compounds.

[55] It should be noted that there are many sources of errors in these simulations (e.g., emissions, lateral boundary conditions for a relatively small domain) and the technique presented here only corrects one of the errors. It should also be noted that the modeling domain was extremely dry during the period of this study. Therefore the impact of inclusion of observed clouds on photochemistry during other periods with more cloud formation could be even more drastic than what was presented in this study. This is evident when the statistical analyses of the results are compared to the large error reduction at the individual sites impacted by clouds. The statistics for the entire domain generally show a moderate improvement. Such large errors can lead to major problems in the use of photochemical models for case studies as well as in air quality forecasting. In case studies, simply an inconsistency between the observed cloud field and that of the model can result in erroneous concentrations that cannot be explained by the in situ measurements. Air quality forecast models often use the model results from the previous forecast (or some adjusted form of it) to initialize the model for the new forecast. Therefore the errors arising from an inconsistency in the cloud fields can propagate into the future forecasts. Therefore the use of observed clouds in the preparation of initial concentrations for air quality forecasting is beneficial.

[56] This study showed that at some locations the errors in ozone concentration arising from inaccurate cloud cover specification reached as high as 60 ppb which was mostly corrected by the use of our technique. Such errors are significant and can have considerable impact on air quality modeling efforts. However, other sources of error in the model due to inadequate cloud specification are as important and need to be addressed. The assimilation technique presented here only corrected the photolysis rates and did not account for the inconsistencies in dynamics and aqueous phase chemistry. The discrepancy in dynamics affects the vertical mixing which can lead to overprediction/underprediction of pollutants. Such discrepancies also affect the chemistry as the heterogeneous processes in the model will be affected by an inconsistent photolysis rate.

[57] These problems require further research for improving the existing photolytic rate calculations in the current air quality models. Even though in this study the technique was implemented within CMAQ, any model that uses model-generated clouds suffers from the same problems and may equally benefit from using satellite-derived clouds. The method presented here addresses a problem in the chemical transport model while the source of this problem is the inadequate cloud prediction in the model. One approach to resolve this issue would be the assimilation of observed clouds in a dynamically consistent manner in the model.

[58] **Acknowledgments.** This work was accomplished under partial support from Texas Commission on Environmental Quality (TCEQ), Cooperative Agreement between the University of Alabama in Huntsville and the Minerals Management Service on Gulf of Mexico Issues, and the following grants: U.S. EPA Star grant R-826770-01-0, Southern Oxidant Study, U.S. EPA Cooperative Agreement R-82897701-0 and Texas Air Research Center/Lamar University contracts TARC/LU-052UAL0030A and 123UAL2030A. Note the results in this study do not necessarily reflect policy or science positions by the funding agencies.

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