IMPROVING CLOUD IMPACTS ON PHOTOLYSIS USING AN ON-LINE RADIATION MODEL IN CAMx

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1. INTRODUCTION

Photochemistry is strongly influenced by the presence of clouds, which can both attenuate and enhance the actinic flux of ultraviolet (UV) radiation responsible for photolysis (Madronich, 1987; Matthijsen et al., 1998; Voulgarakis et al., 2009). The specific radiative impact depends on many factors, including cloud depth, water content, water phase and form (Liou, 1992; Monks et al., 2004).

Clouds are some of the most difficult meteorological phenomena to accurately simulate. As aptly noted by Dolwick (2006) their widely varying spatial and temporal scales are often ill-suited to Eulerian modeling applications, especially at scales addressed by photochemical modeling. Although today's most advanced meteorological models perform well at characterizing large-scale cloud patterns, these models must apply complex parameterizations to account for sub-grid clouds. The simulation of cloud influences at these smaller scales is fraught with uncertainty and is often the source of poor performance (e.g., Yucel et al., 2003). An additional factor for "off-line" or decoupled air quality models is adequately communicating meteorological information from the meteorological model to the photochemical model.

This paper describes updates to The Weather Research and Forecasting (WRF) model (Advanced Research WRF [ARW] core; Skamarock et al. [2008]) and the Comprehensive Air quality Model with extensions (CAMx; ENVIRON, 2010) that address the most important deficiencies in the treatment of cloud impacts on photolysis rates. Details on the formulation of both models are provided in the cited literature. CAMx has treated cloud impacts on clear-sky photolysis rates using a parameterization developed for the Regional Acid Deposition Model (RADM; Chang et al., 1987), a chemical transport model developed in the late 1980's. Since meteorological models such as WRF output only grid-resolved cloud information, accounting for sub-grid cloudiness requires an external diagnostic estimate of sub-grid cloud properties from the grid-resolved thermodynamic parameters output by WRF.

Both WRF and CAMx were updated to improve the characterization of cloud effects on photolysis rates by: (1) outputting explicit sub-grid cloud information from the meteorological model and transferring these data fields to CAMx, and (2) embedding a new fast in-line version of the Tropospheric Ultraviolet and Visible (TUV) radiative transfer model (Madronich, 2002) within CAMx. Using TUV to replace the original RADM treatment provides a more accurate representation of cloud effects on photolysis rates by allowing clouds to be directly involved in the radiative transfer calculations through each grid column. Testing of both WRF and CAMx was conducted for an existing Houston modeling episode developed by the Texas Commission on Environmental Quality (TCEQ).

2. WRF UPDATES

WRF was enhanced to output sub-grid cloud information from the new Grell ensemble cumulus scheme introduced in WRF/ARW v3.0 (Skamarock et al., 2008; Grell and Devenyi, 2002). This vari-

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ant of Grell's cumulus treatment allows subsidence effects to be spread to neighboring grid columns, making it more suitable for finer grid spacing (< 10 km). Not only does a direct pass-through of sub-grid cloud data remove diagnostic guess-work in the process of transferring information to CAMx, but it also enables a more detailed treatment of cloud processes within CAMx. The new output data are processed within the WRF-CAMx interface program to prepare sub-grid cloud water and optical depth fields needed in CAMx for use in the new TUV cloud/radiative treatment. These updates are based on WRF/ARW version 3.2, which was publicly released by the National Center for Atmospheric Research (NCAR) in April 2010. The enhanced version of WRF is referred to hereafter as WRF/Grell v3.2.

3. WRF TESTING

WRF was run in a series of simulations that tested the revised Grell cumulus scheme, primarily to ensure that toggling the output of sub-grid cloud fields did not impact model results. The Grell subsidence spreading option was invoked in all cases. The TCEQ prepared WRF datasets for a 30 August – 7 September 2006 ozone episode that occurred in the area of Houston, Texas. This period was identified specifically because of the high density of cloud cover that occurred over southeast Texas. The TCEQ modeling domain comprises four nested grids with 36, 12, 4, and 2 km spacing, with the finest meshes covering southeast Texas. The WRF physics configuration for all runs was set according to previous TCEQ WRF runs.

Two simulations were run for the August/ September 2006 episode: (1) a WRF/Grell v3.2 run with no radiation feedback in the cumulus scheme ("Run 1"); and a WRF/Grell v3.2 run with radiation feedback invoked ("Run 2"). Surface rainfall, 2-m temperature, and planetary boundary layer (PBL) depth were compared to assess differences among the different WRF versions and configurations.

Noticeable reductions in precipitation, surface temperature, and PBL depth were seen in Run 2, especially in west Texas, directly coinciding with convective activity. This was an expected outcome, since the Grell radiation feedback option accounts for sub-grid cloud influences on the atmosphere's thermal structure. Certainly cloud shading of the ground is shown to cool the surface and lower PBL depths, which feeds back to the convective scheme by removing convective energy and reducing precipitation. We have some concern that the collapsed PBL depths can shut down boundary layer mixing within CAMx, a process that is crucially important to simulate correctly in photochemical models.

The WRF-CAMx interface was run to evaluate the differences in total cloud patterns with the introduction of data output directly from the Grell cumulus scheme. WRF-CAMx was run three ways using output from WRF Run 1: (1) no sub-grid clouds diagnosed or processed; (2) sub-grid clouds diagnosed using the original approach; and (3) sub-grid clouds passed through from the Grell cumulus output fields. Ground-level vertically integrated cloud optical depths over the 12 km grid at 3 PM CST on 1 September 2006 are shown for all three cases in Figure 1. The addition of sub-grid clouds (diagnosed and passed through) added a significant amount of cloud cover over the no sub-grid cloud case during the entire period. The original diagnostic approach tended to generate higher values of cloud optical depth (meaning more cloud water and/or column fractional coverage) relative to the Grell pass-through approach.

4. A STREALINED VERSION OF TUV

The development of efficient radiative transfer models (RTM) such as Fast-TUV (Tie et al., 2005) and Fast-JX (Neu et al., 2007) has made it feasible to embed such models as in-line algorithms within photochemical grid models. These in-line RTMs directly calculate photolysis rates while interactively accounting for the evolution of clouds and aerosols throughout the domain. To be sufficiently economical, several simplifications must be made in these RTMs, such as reducing the number of wavelength bands and using plane-parallel two-stream approximations.

We are concerned with the degree to which these simplifications broaden uncertainty in the photolytic reaction rates and yields. Therefore, our approach centered on using a streamlined version of TUV within CAMx for the single purpose of calculating cell-specific cloud adjustment factors (replacing the RADM parameterization), which are directly applied to predetermined clear-sky photolysis rates. The clear-sky photolysis rates continue to be derived externally (using the full-science TUV) and passed to CAMx in a lookup table format.

The in-line TUV is run for each cloudy grid column in a manner that yields a vertical profile of cloud-to-clear (cloud:clear) actinic flux ratio by

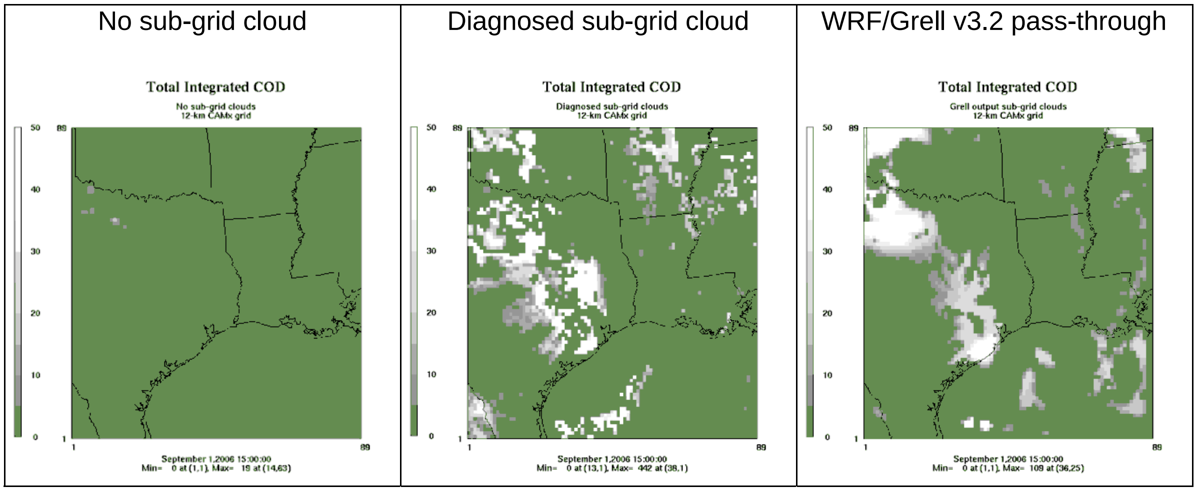

Figure 1. Surface total vertically integrated cloud optical depth on the 12 km CAMx grid at 3 PM CST, 1 September 2006 for the no sub-grid cloud case (left), diagnosed sub-grid cloud case (middle), and WRF/Grell v3.2 pass-through case (right). Cloud fields were derived from WRF Run 1.

layer, which is then applied as a multiplicative factor to the clear-sky values extracted from the lookup table. This approach maintains accuracy in the calculation of clear-sky photolysis rates, while allowing clouds to be directly involved in radiative transfer calculations through each grid column.

TUV was substantially streamlined. First, radiative flux calculations are performed for only a single representative wavelength (350 nm) to minimize runtime requirements. According to Monks et al. (2004) there is little systematic difference during midday hours in how clouds affect the photolysis rates of NO_2 and O^1D, two compounds with very different action spectra. We independently confirmed this with TUV. Second, since absorption by gases occurs in rather narrow UV bands relative to the broad-band influence of clouds, the absorption from oxygen, ozone, nitrogen dioxide and sulfur dioxide were removed. Third, the extraterrestrial flux was not needed as it cancels out in the calculation of the cloudy:clear ratio; this removed a significant amount of computation associated with wavelength interpolations. Finally, the plane-parallel version of the delta-Eddington approach was used in lieu of the more complex and expensive pseudo-spherical geometry.

The current implementation of this streamlined TUV ignores effects of aerosols, but this capability is included as a place-holder for future updates. Aerosol optical depth is included in the clear-sky photolysis calculations performed in the full science TUV pre-processor.

Preliminary tests against the full-science TUV showed that the streamlined version run outside of CAMx resulted in less than 1% differences in cloudy:clear actinic flux ratio. Timing tests of the streamlined TUV showed that the impact to CAMx run time should typically be on the order of one second per simulation hour.

5. CAMx MODIFICATIONS

CAMx was modified to include the fast TUV model to calculate and pass back photolysis adjustment factor profiles through each grid column containing at least one cloudy layer. TUV is not called for completely clear grid columns, and the adjustment profile in such cases is set to a uniform value of 1. The original RADM cloud adjustment approach was retained as an option. Both the original RADM and new TUV photolysis adjustments are calculated using the cloud optical depth fields supplied by the WRF-CAMx interface. The fast TUV solver is called each time the meteorological data are updated (usually at the top of each hour). Unlike other meteorological fields such as temperature, pressure, and mixing rates, the cloud fields are held static between update times since they are processed to represent time-averaged fields. The static hour-averaged cloud fields carry the most influence on the resulting cloud adjustment profiles (as opposed to solar zenith angle or atmospheric density), so limiting TUV calls to the meteorological update cycle minimize impacts to runtime. Other meteorological parameters such as temperature and pressure are averaged between update times for use in TUV.

Like previous versions of CAMx, a lookup table of clear-sky photolysis rates must be generated using the full-science TUV pre-processor. This version of TUV provides a very accurate estimate of photolysis rates by integrating over many UV wavelengths, and applying a much more comprehensive solver technique. The look-up table contains photolysis rates for a range of solar zenith angles, altitudes, surface albedo, ozone column, and haze optical depths. This table is read by CAMx at the start of the simulation (as in previous versions), and the appropriate photolysis values are extracted and interpolated from the table according to conditions in each grid cell at each time step. The TUV or RADM cloud adjustment factors are then applied to the photolysis rates just before the chemical mechanism solver is called.

6. CAMx TESTING

A series of CAMx simulations was conducted to test the impacts of: (1) the original RADM cloud adjustment vs. the in-line TUV adjustment; and (2) the WRF/Grell v3.2 sub-grid cloud pass-through vs. the original sub-grid cloud diagnosis. CAMx was run for the 31 August – 7 September 2006 Houston modeling episode; the model configuration and non-meteorological input data were provided by the TCEQ, while the meteorological inputs were developed from WRF Run 1 (described above).

The following CAMx simulations were run for the entire modeling period:

- Run 1 (RADM/GSGC): RADM cloud adjustment, Grell-derived sub-grid clouds;
- Run 2 (TUV/GSGC): TUV cloud adjustment, Grell-derived sub-grid clouds;
- Run 3 (TUV/DSGC): TUV cloud adjustment, diagnosed sub-grid clouds;

Impacts to hourly surface ozone were evaluated since cloud impacts tend to be on short time scales, and effects would be less apparent for 8-hour ozone.

Figure 2 displays daily peak 1-hour surface ozone on the 12 km grid for Run 2 (TUV/GSGC), as well as the difference between Run 2 and Run 1 (RADM/GSGC) on 5 September 2006. Daily peak ozone on 5 September consistently exhibited the largest impacts among all episode days. Overall, the TUV adjustment resulted in a slight reduction in ozone relative to the RADM adjustment. The largest reductions rarely exceeded 10 ppb, and were grouped under transient cloudy areas. Some slight ozone increases are seen, but rarely did they exceed 1-2 ppb. A frontal band traversed the area from north to south during the middle of the episode, with the most dramatic surface ozone effects on 5 September. Generally wide areas of slight ozone reduction occurred with the introduction of the TUV cloud adjustment, and therefore ozone sensitivity to the choice of TUV or RADM was not large for this episode.

We further investigated model sensitivity to the approach by which sub-grid clouds are generated. Run 3 used the TUV cloud adjustment, but sub-grid clouds were diagnosed (DSGC) according to the original approach in WRF-CAMx. Figure 3 displays the difference between Run 3 and Run 2 for daily peak 1-hour surface ozone on the 12-km grid. Ozone results were much more sensitive to the amount of cloudiness passed to CAMx than the approach by which cloud photolysis adjustments are applied.

On each day, Run 3 resulted in large local ozone differences (both positive and negative) of up to several tens of ppb. This reflects the different locations, lifetimes, and cloud densities between the two sets of cloud inputs. Run 3 showed a slight tendency for larger and more widespread ozone increases than decreases, indicating that the pass-through of Grell cumulus cloud data tended toward more cloudiness spatially and/or temporally than the diagnostic option. Note especially on 5 September, that the diagnostic option removed much of the Grell-derived cloudiness along the frontal band extending from Dallas eastward into southern Arkansas.

7. SUMMARY AND CONCLUSIONS

Clouds play an important role in photochemical simulations, and their specific impact depends on their varied physical characteristics. Even with today's modern and sophisticated meteorological models, clouds remain one of the most difficult meteorological phenomena to accurately simulate, especially at sub-grid scales. As a result, uncertainties in cloud fields can translate to large errors in photo-chemical models. Furthermore, the most common meteorological models used to drive "off-line" photochemical models do not output important sub-grid cloud variable fields.

We have described an improvement to WRF and CAMx that involves: (1) transferring explicit sub-grid cloud information from the meteorological model, and (2) embedding an in-line version of TUV within the photochemical model. The WRF-CAMx interface program was modified to utilize the new Grell cumulus output variable fields in order to develop improved sub-grid cloud and precipitation fields for the purpose of calculating cloud optical depths, which are passed through to

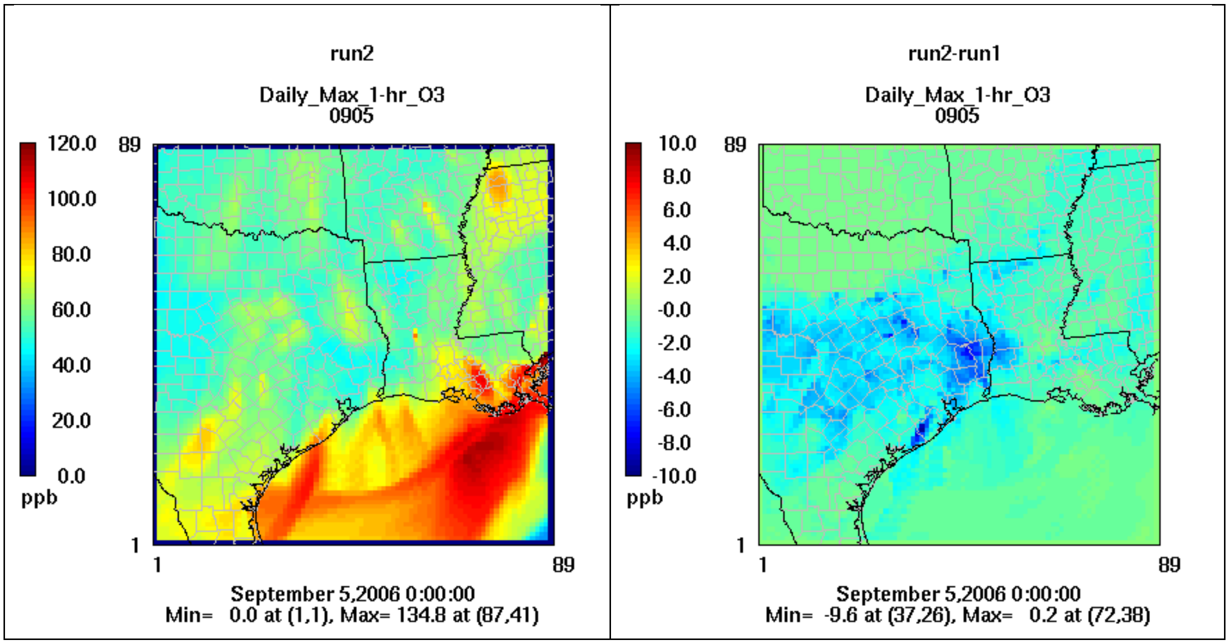

Figure 2. Daily maximum 1-hour ozone in the 12 km TCEQ CAMx grid on 5 September 2006 for Run 2 (left), and difference between Run 2 and Run 1 (right).

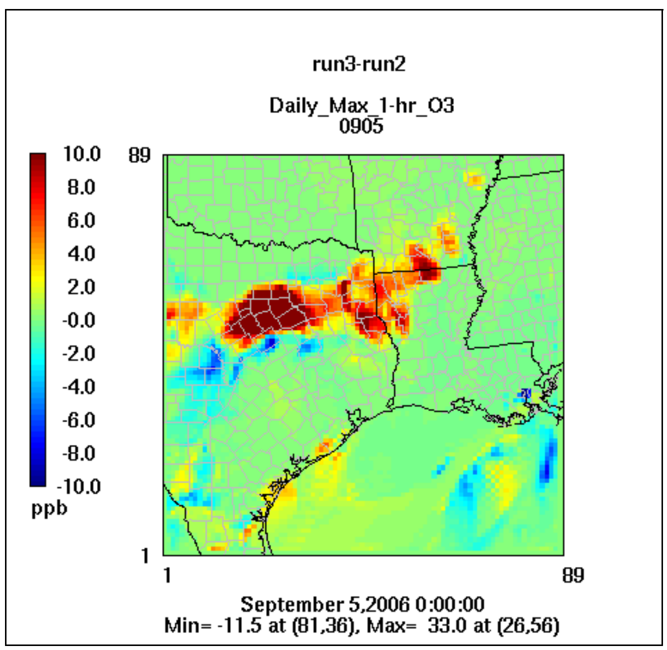

Figure 3. Difference in daily maximum 1-hour ozone (Run 3 – Run 2) in the 12 km TCEQ CAMx grid on 5 September, 2006.

CAMx. WRF-CAMx retains the option to diagnose sub-grid cloud fields if the Grell cumulus option is not available.

A series of WRF and CAMx simulations over the August/September 2006 Houston episode was run to test the updated modeling system and impacts to hourly surface ozone from various cloud processing methodologies. Based on the model updates, analyses, and findings from the activities described herein, we have identified several elements for future work. First, the system will be tested and evaluated for a wider range of cloud and photochemical environments. Second, aerosol influences, different cloud water phases, and dynamic surface UV albedo (e.g., to account for time-evolving conditions such as snow cover) will be added to the in-line TUV calculations. Third, we will investigate ways to minimize impacts to vertical mixing processes that may occur when PBL depths collapse under convective activity when the Grell cumulus-radiation feedback is invoked. Finally, we will further investigate whether increasing the number of wavelength bands used by the in-line TUV would significantly improve accuracy.

8. ACKNOWLEDGEMENTS

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9. REFERENCES

Chang, J. S., R. A. Brost, I. S. A. Isaksen, S. Madronich, P. Middleton, W. R. Stockwell, and C. J. Walcek, 1987: A three-dimensional Eulerian acid deposition model: Physical concepts and formulation. *J. Geophys. Res.*, **92**(D12), 14,681–14,700.

Dolwick, P., 2006: The effects of cloud attenuation on air quality: A comparison of model treatments. *Proc. 14th Joint Conference on the Applications of Air Pollution Meteorology with the Air and Waste Management Association*, Atlanta, GA, American Meteorological Society.

ENVIRON, 2010: Users Guide: Comprehensive Air quality Model with Extensions (CAMx), Version 5.20 (April, 2010). ENVIRON International Corporation, Novato, Califorina, [Available online at www.camx.com.]

Grell, G. A., and D. Devenyi, 2002: A generalized approach to parameterizing convection combining ensemble and data assimilation techniques. *Geophys. Res. Lett.*, **29**, 1693-1696.

Liou, K. N., 1992: Radiation and cloud processes in the atmosphere, theory, observation, and modeling. *Monogr. on Geol. and Geophys*., No. 84, edited by R. Dmowska, J. R. Holton, and H. T. Rossby, Oxford Univ. Press, New York.

Madronich, S, 1987. Photodissociation in the atmosphere 1. Actinic flux and the effects of ground reflections and clouds. *J. Geophys. Res*., **92**, 9740–9752.

Madronich, S., National Center for Atmospheric Research, Boulder, Colorado, cited 2002: Tropospheric ultraviolet and visible radiation model. [Available online at http://acd.ucar.edu/models/UV/TUV/index.html.]

Matthijsen, J., K. Suhre, R. Rosset, F. L. Eisele, R. L. Mauldin, and D. J. Tanner, 1998: Photodissociation and UV radiative transfer in a cloudy atmosphere: Modeling and measurements. *J. Geophys. Res*., **103**(D13), 16665–16676.

Monks, P., A. Rickard, and S. Hall, 2004: Attenuation of spectral actinic flux and photolysis frequencies at the surface and through homogeneous cloud fields. *J. Geophys. Res.*, **109**, D17206, doi:10.1029/2003JD004076.

Neu, J., M. Prather, and J. Penner, 2007: Global atmospheric chemistry: Integrating over fractional cloud cover. *J. Geophys. Res*., **112**, D11306, doi:10.1029/2006JD008007.

Skamarock, W. C., J. B. Klemp, J. Dudhia, D. O. Gill, D. M. Barker, M. G. Duda, X.-Y. Huang, W. Wang, and J. G. Powers, 2008: A description of the Advanced Research WRF version 3. NCAR Tech. Note NCAR/TN-45+STR, National Center for Atmospheric Research, Boulder, Colorado. [Available online at http://www.mmm.ucar.edu/wrf/users/.]

Tie., X., S. Madronich, S. Walters, R. Zhang, P. Rasch, and W. Collins, 2003: Effects of clouds on photolysis and oxidants in the atmosphere. *J. Geophys. Res.*, **108**, D20, doi:10.1029/2003JD003659.

Voulgarakis, A., N. H. Savage, O. Wild, G. D. Carver, K. C. Clemitshaw, and J. A. Pyle, 2009: Upgrading photolysis in the p-TOMCAT CTM: model evaluation and assessment of the role of clouds. *Geosci. Model Dev.*, **2**, 59-72, [Available online at www.geosci-model-dev.net/2/59/2009/.]

Yucel, I., W. J. Shuttleworth, X. Gao, and S. Sorooshian, 2003: Short-term performance of MM5 with cloud-cover assimilation from satellite observations. *Mon. Wea. Rev*., **131**, 1797-1810.