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## AN ANALYSIS OF THE IMPACTS OF GLOBAL CLIMATE AND EMISSIONS CHANGES ON REGIONAL TROPOSPHERIC OZONE

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#### ABSTRACT

Many of the synergistic impacts resulting from future changes in emissions as well as changes in ambient temperature, moisture, and uv flux have not been quantified. A three-dimensional regional-scale photo-chemical model (STEM-II) is used in this study to evaluate these perturbations to trace gas cycles over the eastern half of the United States of America. The model was successfully used to simulate a regional-scale ozone episode (base case - June 1984) and four perturbations scenarios - viz., perturbed emissions, temperature, water vapor column, and incoming UV flux cases, and a future scenario (for the year 2034). The impact of these perturbation scenarios on the distribution of ozone and other major pollutants such as SO<sub>2</sub> and sulfates were analyzed in detail. The spatial distribution and the concentration of ozone at the surface increased by about 5-15% for most cases except for the perturbed water vapor case. The regional scale surface ozone concentration distribution for the year 2034 (future scenario) showed an increase of non-attainment areas. The rural areas of Pennsylvania, West Virginia, and Georgia showed the largest change in the surface ozone field for the futuristic scenario when compared to the base case.

#### INTRODUCTION

A growing concern over global climate change has been instrumental in focusing the scientific community's attention toward the need to study its varied impacts. Issues pertaining to global warming, loss of stratospheric ozone, and their respective influences on human activities and the surrounding environment have become more important topics of research. Recent studies1 seem to focus on the impacts of trace gases on possible climate change. There are, however, limited studies that address the issue of the impacts of a possible climate change scenarios on the trace gas distribution of the troposphere. Under a changed climate scenario, the tropospheric trace gas distribution would significantly alter. An increase in surface temperature (corresponding to global warming) would cause an increase in the water vapor concentration in the atmosphere. An increased water vapor concentration would then amount to an increase in the  $H_2O_2$  and OH concentrations and a subsequent decrease in the ozone concentration field.<sup>2</sup> This would, thus, affect the oxidative capacity of the atmosphere. Moreover, due to anthropogenic and/or natural causes the atmospheric composition would change over a period of

time. Such changes in atmospheric levels of certain pollutants (NOx, NMHC, CO, etc.) have the potential to affect the rate of tropospheric ozone formation and the abundance of the free radical oxidant OH on a global scale. Perturbations to stratospheric O<sub>3</sub> and climate (temperature and moisture) can also influence the rates of key photochemical processes affecting tropospheric O<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, and OH. Since these species are the principal tropospheric oxidants, the above mentioned changes may alter the overall oxidizing capacity of the atmosphere.

A few studies<sup>3,4,5</sup>, using one-dimensional models, have attempted to better understand these impacts on the complex tropospheric chemistry. Such studies, however, are limited and their authors recommend that the sensitivities/ changes should be evaluated using a three-dimensional model because  $O_3$  is sufficiently long-lived to have a widespread effect on the tropospheric chemistry.

The impact of various perturbations due to global climate change coupled with changes in emissions can be studied in detail using a comprehensive regional-scale photochemical model. In this paper, the author will make an attempt to address various issues pertaining to these climate and emission change attributes and their impact on the tropospheric trace gas distribution by utilizing STEM-II photochemical model.<sup>6</sup>

#### STEM-II APPLICATION

For a detailed study of the possible perturbations to the trace gas distribution over eastern U.S.A. due to the anticipated changes in global climate attributes and the regional-scale emissions, a three-dimensional regional-scale comprehensive photochemistry model (viz., the STEM-II)6 was used in this study. The modeling domain comprised of 22x28x11 grid points and the horizontal grid spacing was 80 km and the vertical resolution being 500m. This study domain covers the entire eastern United States. The 1985 NAPAP emissions inventory 7 was used in this study. The episode selected as the base case was clear-sky summer-time conditions on 7th - 11th June, 1984.8 During this period, the entire modeling domain was under the influence of a high pressure system centered over the Atlantic Ocean off the coast of South Carolina. Most of the region was under clear sky during the event. This was a typical high-pressure dominated stagnant summer event over eastern United States that results in high photochemical oxidant episodes. Shin<sup>8</sup> provides a detailed description of the grid setup, emissions and the meteorological inputs.

In order to simulate a future scenario for the year 2034, large number of gas-phase-only simulations were performed using meteorological and emissions data for the base year of 1984. Table 1 shows the different scenarios simulated to study the impact of perturbations to key parameters on regional-scale trace gas distribution. The climate scenarios are consistent with present climate change scenarios (Intergovernmental Panel on Climate Change),9 while those for the emissions are estimated from EPA,10 OTA,<sup>11</sup> and Kavanaugh.<sup>12</sup> There, however, remains a large uncertainty in the projections of future NOx and NMHC emissions. Each simulation consisted of a 48-hour run for initialization using the meteorological inputs of June 5-6, 1984. This provided us with the initial distributions consistent with the emissions and meteorological fields. Following this, a 5-day (108 hours) simulation was conducted and the results analyzed over a broad spectrum of pollutant species and their behavior. The impact on the trace gas distribution of all perturbation scenarios listed in Table 1 are analyzed.

Table 1 : List of Scenarios Simulated

- (a) Base Case with biogenic emissions and diurnal dry deposition incorporated
- (b) Perturbed Emissions scenario for the year 2034 : 33% increase in NOx during 1984-2034 due to an increase in automobiles/highway miles and a simultaneous increase in stationary sources.
   25% decrease in SOx corresponding to a realistic decrease in sulfur sources subject to

the 1990 Clean Air Act requiring a 50% cut

- in sulfur emissions from power plants.
  (c) Perturbed Surface Temperature and Vertical Temperature profile scenario approx. a 3K change in surface areas for the mid-latitudinal areas <sup>13</sup>
   (d) Perturbed water vapor scenario approx. 20 - 27% increase in the lower troposphere corresponding to an increase of surface temperature of 3 K <sup>2</sup>
- (e) Perturbed uv flux scenario approx. a 10% increase in uv flux was accounted for the decrease in the stratospheric ozone
   (f) Total Perturbation scenario - Overall projected
- (f) Total Perturbation scenario Overall projected scenario for the year 2034 from a combination of cases (b) through (e)

## **RESULTS AND DISCUSSION**

The STEM-II model was successfully used to simulate an actual episode (base case - year 1984), a future scenario (year 2034), and four perturbation scenarios to test the sensitivity of tropospheric trace gas distribution to perturbations to global climate attributes and emissions change. The trace gas distribution of selected species (including ozone) were studied in detail and the impacts fully analyzed for all the above mentioned cases.

The spatial distribution of ozone at the surface increased for most cases except for the perturbed water vapor case. Figures 1(a) through 1(e) represent the percent change of ozone concentrations at the ground level, for each perturbation case, over the entire study domain. In the case of perturbed emissions, (see Figure 1a), the ozone concentrations increased by about 5-10% over the eastern However, the upper midwest regions land mass. encompassing Indiana, Michigan, and Ohio, and the urban centers in New Jersey and New York showed an actual decrease in ozone (-5 to -15%) at the surface. This can be explained by the already low hydrocarbon/NOx ratios in those regions. By increasing the NOx values to even higher levels, while the tropospheric hydrocarbon levels remain constant due to low biogenic sources and constant anthropogenic emissions, puts the photochemistry in the regime where formation of nitric acid (terminating reaction) becomes an important sink for all available HOx. Consequently, any further increase in NOx decreases the urban ozone concentrations.

In the case of perturbed temperature (+3 K) scenario (see Figure 1b), where the emissions were retained to the base levels, the regional-scale ozone concentration increased by about 5 - 15% over most of the domain. The effect of temperature on ozone is primarily due to the effect on the lifetime of PAN and its homologues.

In the case of perturbed water-vapor case (see Figure 1c), where the emissions and temperatures were retained to the base levels, the regional-scale ozone showed very little change. An increase in water vapor suggests an increase in the hydroxyl radical pool and a decrease in the ozone concentration.

In the case of an increase in the UV flux ( $\sim 10\%$ ) (see Figure 1d), the regional ozone values increased by about 5-15%. This is accounted primarily by the daytime photochemistry and its subsequent carryover of higher values into the night time.

In the event of the total perturbation scenario (see Figure 1e), where all of the above perturbations were accounted to hypothetically simulate the year 2034, overall increase of surface ozone concentration was noted to be in the range of 10-25%. This implies an increase in the number of the non-attainment areas within the study domain.

The regional-scale surface ozone concentration predicted for the year 2034 (viz., total perturbation case) showed an increase of non attainment areas with the greatest changes observed in certain rural areas including the ones in Pennsylvania, West Virginia, and Georgia. Figure 2 highlights this feature by comparing the total perturbation case (2034) to the base case.

The diurnal variations of the ozone cycle remained similar and the differences between rural and urban profiles were essentially retained in all cases. Within the lower troposphere, the ozone profile showed a large change for the perturbed emissions and the total perturbation case. Rural values of ozone were most affected by these perturbations.

Two points were selected within the study domain representing an urban and a rural area in Georgia. Figure 3 (a and b) gives the percent change in the ozone concentrations at the ground level, for each perturbation with respect to the base case, at these two sites. The percent change over the base case is computed for a 24-hour period. As seen in the graph, the surface values of ozone in the rural Georgia area increased significantly for the total perturbation case. Increase in the temperature also had a large effect on the ozone values. The other perturbations caused small increases in the ozone concentrations. However, the changes in the ozone concentrations in the urban Georgia site were relatively smaller. The constraint here could be the low hydrocarbon/NOx ratio in the urban regions when compared to higher hydrocarbon/NOx ratios in the rural areas.

In conclusion, this study has demonstrated that significant changes in trace gas and photochemical oxidant cycles can be anticipated under global climate change scenarios. Increases of upto 40% in ozone concentrations are predicted as a direct impact of changes in climate attributes and emissions. Such increases in tropospheric ozone could possibly have feedback effects on the regional climate. Rural areas and areas downwind of pollution hotspots would experience significant changes in the tropospheric composition. Regional-scale photochemical models, such as STEM-II, serve as invaluable tools in detailed studies of tropospheric trace gas distribution. This brief introduction to the sensitivity of trace gas distribution to global climate change and emission change should provide a basic insight into similar research endeavors.

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