Review of Regional evaluation of the performance of the global CAMS chemical modeling system over the United States (IFS cycle 47r1) by Williams et al.

General summary

This paper updated the homogeneous and heterogeneous NOx chemistry applied in the three independent tropospheric-stratospheric chemistry modules maintained within CAMS and evaluate lower tropospheric O3, CO, and NO2 over the contiguous US. The developments are evaluated against multi-platform observations and the chemically induced biases in different species is estimated to be within 10 ppbv. The authors find large model spread between chemistry schemes during winter in lower tropospheric ozone (10-35%) and oxidative capacity which is attributed to NOx lifetime differences. The study concludes that chemically induced differences in quality of CAMS forecasts over CONUS vary by season, species, altitude, and region. The intercomparison of different chemistry configurations is interesting but I find several major weaknesses in the paper as listed below.

First, I was expecting that the authors will evaluate the impact of the updates to the homogeneous and heterogeneous chemistry against the older model version without these updates to demonstrate how important these updates here. However, such a comparison is not presented. Thus, the real value of these updates is not quantified.

Second, the authors compared the three chemistry configurations against the CAMS reanalysis (CAMSRA). At lines 76-77, the authors state that any changes in the operational system should ensure that the change improves the model performance with respect to observations. In most cases (e.g., Figures 4-6), the performance of CAMSRA is better than all the three individual configurations. Since any of the new configurations here do not outperform CAMSRA, does it mean that these configurations may not be suitable for operational adaptation? Furthermore, the assimilation of several species in CAMSRA makes a direct comparison of three chemistry simulations with CAMSRA difficult.

Third, several discrepancies between the observations and model simulations are not clearly explained (see specific comments for details).

Fourth, the choice of US as a polluted region over Asia is not motivating enough given that anthropogenic emissions in Asia are much higher than the US.

Therefore, I recommend major revisions of the paper before it can be considered for publication in GMD. Below are some specific comments.

Specific comments

Lines 57-58: Operational air quality forecasts over the US are not only provided by the CAMS system. The US has their National Air Quality Forecasting Capability (NAQFC) that provides air quality forecasts at 12 km grid spacing twice per day and the NAQFC forecasts are used by

decision-makers to issue advance notices and warnings for air pollution episodes. Since lines 43-55 discuss aspects and benefits of air quality forecasting, it is relevant to discuss NAQFC here.

Lines 63-64: Anthropogenic emissions in the US are much lower than Asia and thus it is hard to see this as a motivation for focusing on the US.

Line 99: To what heterogeneous reactions you are referring here?

Line 105: I suggest replacing the term "mini-ensemble" with "multiple chemistry simulations".

Line 144: Do all the chemistry configurations use the same quantum yield and cross sections for calculation of photolysis frequencies? Also, how are the effect of aerosols treated in photolysis rate calculation? If not, can the 5% differences in photolysis frequencies be attributed to these factors?

Tables 2-5: Please include a description of various symbols (Ko, R, T, and QY etc.) used in the rate expression column in the captions of these table.

Line 168: Do you provide HONO emissions to the model as well?

Tables 3-5: Since you are not validating the updated isoprene, aromatic, HCN, and CH3CN chemistry, I think Tables 3-5 can be moved to supplementary material.

Line 442-443: The minimum is not centered around Colorado but rather is spread over Oklahoma, Nebraska, South Dakota, North Dakota, and parts of Texas.

Line 446: Can you explain why the decrease in MOZ is due to reduced transport? What are the source regions affecting the surrounding oceans?

Fig S1: The differences in OH are interesting. While it makes sense that higher ozone can lead to higher OH levels through photolysis but I am unbale to understand why OH increase in summertime in MOZ over the western US despite of a decrease in ozone mixing ratios. Any thoughts?

Figure 4: The observed ozone seasonal cycle at NWR looks suspicious. Most of the sites in Colorado show a summertime maximum. I looked at multi-year hourly ozone time series from NOAA GML website (see below) and found that there are a lot of measurement gaps at NWR during 2014. So, I recommend revisiting the data processing and evaluation at this site and revising the corresponding text accordingly.
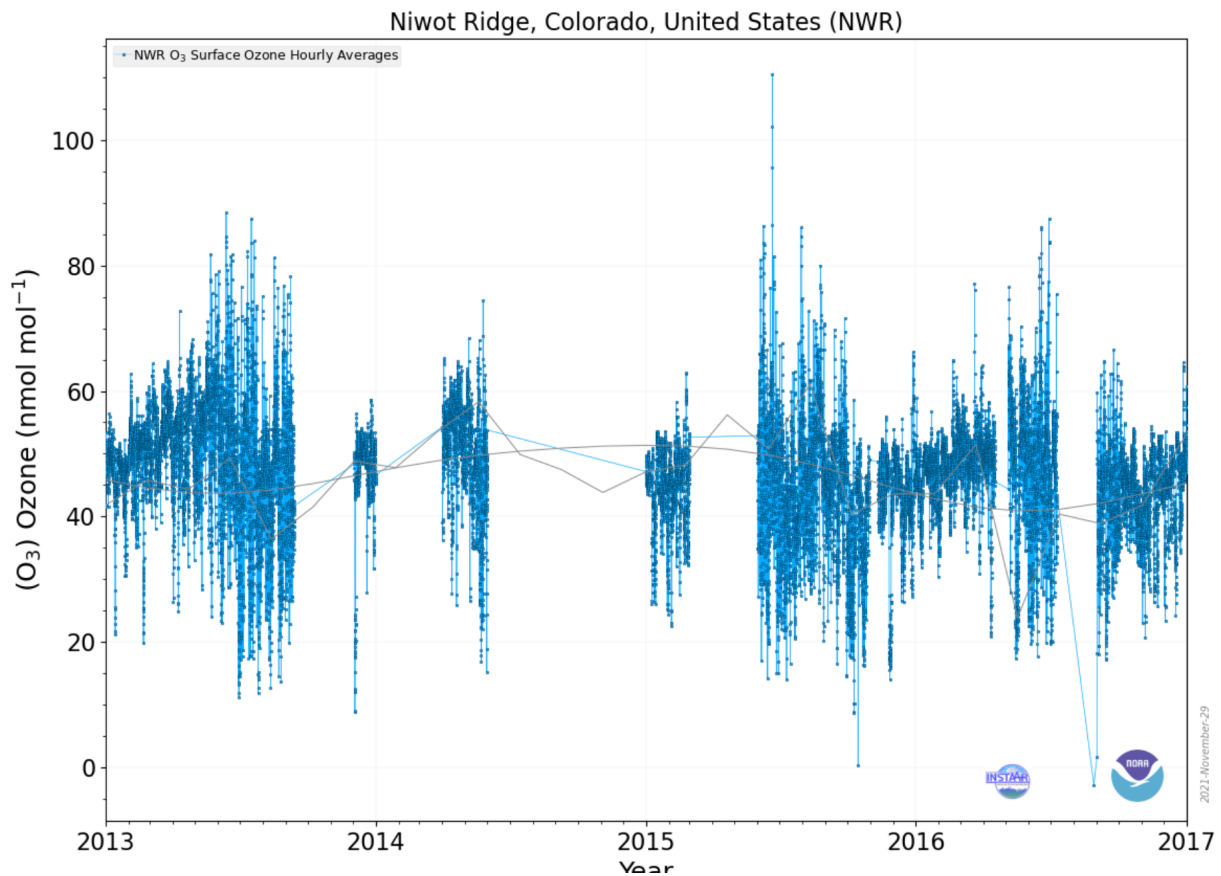
Figure 5: None of the model configurations capture ozone seasonal cycle at rural East Coast sites. All the models predict a summertime peak while the observations show continuous decrease. This is really surprising. Can you throw some light on this?

Lines 535-540: Here you use ppb as ozone units while all the earlier discussion is in micrograms/m3. Please use consistent units.

Line 581-582: Can you provide more insight into the different seasonal cycles of CO between the model and observations at NWR?

Lines 775-783: This is a good discussion but it would have been great to perform a couple of sensitivity experiments using just one of the chemistry configurations to understand the impact of conversion efficiency on N2O5 hydrolysis.