Executive Summary

Tropospheric ozone is produced in the planetary boundary layer through a series of non-linear reactions between oxides of nitrogen (NO_x) and Volatile Organic Compounds (VOCs). Reliable emissions inventories are key to accurate ozone modeling. NO_x concentrations are known to respond quickly to emission changes due to the very short lifetime of NO_x in the atmosphere and a linear relationship with emissions. NO_x emissions from mobile and point sources have significantly decreased over time due to improvement in fuel technology and effective emissions control technologies and strategies. These reductions are well corroborated by satellite and in-situ observations. These emissions reductions imply that the National Emission Inventory (NEI) used in model simulations needs to be updated for use to represent current emissions. A robust procedure to update the emission inventory is to use additional NO_2 observations and a 3-D modeling framework with a realistic representation of sensitivity of NO_x levels to emissions. These improvements can be achieved using the inverse modeling technique which is useful when a direct quantity (here, emissions) is not measurable.

The concept of an inverse problem involves using physical laws to back-calculate the quantity of interest ("a posteriori" emissions) from known or partially-known approximated states ("a priori" ambient measurements or satellite retrievals). Updating emission inventories using satellite measurements is a common application for which remote sensing measurements are cost-effective. Due to spatial inhomogeneity in distributions of NO_x concentrations and limited number of monitoring sites, satellite remote sensing of tropospheric NO_2 is widely used for this purpose. The biggest advantage of using satellite data is the high spatial coverage that makes it possible to update emission inventories over large domains. The National Aeronautics and Space Administration (NASA) records this data through its Ozone Monitoring Instrument (OMI) aboard the Aura satellite; tropospheric NO_2 concentrations from this instrument (Level 2, V2.1) were used for this project. Several adjustments were required to remove noisy observations and re-grid them for our model simulation domain. These included corrections for cloud fraction, quality flags, solar angle and removal of a priori profile influences. Our results revealed that removal of a priori profile influences is crucial to obtaining accurate results. Disregarding this step leads to under-prediction and over-prediction of NO_2 in urban and rural regions respectively.

In this study, a Bayesian inversion of tropospheric OMI NO_2 is conducted to update four source categories of the National Emission Inventory (NEI) 2011 (area, soil, mobile and point sources), as well as the total NO_x inventory. Using a "Brute Force" method, the adjusted NEI-2011 demonstrated an overall reduction in anthropogenic source categories (i.e., area, mobile and point) and an increase in biogenic soil emission of NO_x. The largest reduction in NO_x emission for the area source category was predicted in the center of Houston and Lake Charles city. A large increase in the biogenic NO_x emissions (50%) was found in most rural areas. Additionally, the results indicated that mobile NO_x emissions reduced by 30-40% in urban regions. The largest decrease was found for the center of Houston. The largest decrease for point source category was found for Lake Charles and Houston Ship Channel, ranging between 40-65%. The considerable decrease was mainly due to strict emissions regulations. A new simulation was performed using the United States Environmental Protection Agency (USEPA)'s Community Multiscale Air Quality (CMAQ) model using the updated total NO_x emissions. The model simulation results were evaluated using in-situ data from independent aircraft studies and the Texas Commission on Environmental Quality (TCEQ)'s Continuous Ambient Monitoring Stations (CAMS)

network. Mean error and root mean square error (RMSE) of the model-measurement comparisons were reduced with the updated NO_x emissions. RMSE and bias between aircraft NO_x measurements and simulated ones are 2.4 and 6.0 ppbv for the default NEI-2011 and 1.9 and 4.1 ppbv for the updated NEI-2011. The model-measurement comparisons using CAMs data indicated that, mean absolute bias and RMSE decrease by 0.8 and 1.1 ppbv from the default NEI-2011 to the inverse modeling updated case.

In order to investigate the impact of NO_x emissions changes on chemical conditions (i.e., NO_x-saturated or –sensitive), we calculated the $HCHO/NO_2$ ratio before and after using inverse modeling. A comparison of P3-B aircraft HCHO levels to simulated values demonstrated under predicted HCHO levels likely resulting from biogenic model used. We did not find any difference in HCHO concentrations before and after using inverse modeling due to marginal indirect impacts of NO_x on HCHO. After updating NO_x emissions using inverse modeling, most of the urban regions became increasingly NO_x-sensitive (20-85% increases in the ratio) due to large decreases in anthropogenic NO_x emissions. On the other hand, rural regions showed a transition from NO_x-sensitive toward more NO_x-saturated due to an increase in soil NO_x emissions.

We also investigated the impact of the updated inventory on ozone predictions. Overall, the ozone changes were small both on surface and aloft, suggesting that the Houston region is NO_x saturated. We found statistics (before and after updating emission inventory), for correlation (0.74, 0.76), the Index of Agreement (IOA) (0.79, 0.80), RMSE (14.6, 14.4), MAB (12.0, 11.7) and MB (9.3, 9.3). This is in agreement with previous findings suggesting that other parameters such as VOC emissions and dry depositions should be constrained as well. Small increases in ozone concentrations were observed in NO_x-sensitive rural areas, consistent with the increase in NO_x in the new inventory. Additionally, we applied Objective Analysis (OA) to meteorological modeling using the Weather Research and Forecasting (WRF) model to constrain the modeled fields using in-situ data. The results indicated improvement of the simulated meteorology fields which provides a solid base for subsequent emission and chemistry modeling. Regarding the wind fields which are critical to pollutant transport, the IOA improved by 6% for U-wind and 11% for V-wind after using OA. For temperature, the correlation coefficient increased by about 10% while IOA rose by 8%.